

LA-UR-17-31465

Approved for public release; distribution is unlimited.

Title: Gas composition observed by destructive examination of 3013 containers

Author(s): Veirs, Douglas Kirk
Kelly, Elizabeth J.
Berg, John M.
Nguyen, Binh
McClard, James W.
Hensel, Steve J.
Duffey, Jonathan M.
Scogin, John H.

Intended for: Report

Issued: 2018-03-09 (rev.1)

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Gas composition observed by destructive examination of 3013 containers

D. Kirk Veirs, LANL
Elizabeth J. Kelly, LANL
John M. Berg, LANL
Binh Nguyen, SRS
James W. McClard, SRS
Steve J. Hensel, SRS
Jonathan M. Duffey, SRNL
John H. Scogin, SRNL

For: MIS Program

Date: December 20, 2017

Abstract.

The Department of Energy's Integrated Surveillance and Monitoring Program implements requirements to measure gas pressure and gas composition during destructive examination of 3013 containers as required by DOE's 3013 Standard. Gas pressures and gas compositions measured by the Program during destructive examination of 3013 containers from 2007 to 2017 are reported here. The maximum gas pressure within the population of containers that have undergone destructive examination was 299 kPa (43.4 psia) and this container had 0.52 wt% moisture which is higher than the Standard allows. The majority of containers have essentially no gas pressurization. Hydrogen was quantifiable above the measurement uncertainty in less than half (47 out of 114) of the containers that have undergone destructive examination. The amount of hydrogen that is generated within the sealed 3013 containers is always less than the amount predicted by the 3013 Standard's pressure equation with the maximum observed hydrogen fraction (observed moles of hydrogen/predicted moles of hydrogen) of 0.18. It is recommended that for the purposes of calculating a maximum hydrogen pressure using the 3013 equation, a correction to the amount of hydrogen produced from decomposition of the water be incorporated. The pressure (or moles) of H₂ should be reduced by multiplying by a hydrogen fraction as described in the text. Helium and nitrogen are the other major constituents. Oxygen which is initially present in nearly all of the containers undergoing destructive examination was consumed to low values in all cases. The highest observed oxygen concentration was 1.7%, and in this case the H₂ concentration was essentially zero. Other minor gases observed in some cases included CO₂ with maximum content of 3.5%, CO with maximum content of 0.8%, N₂O with maximum content of 1.4%, and CH₄ with maximum content of 0.4%.

Contents

Abstract	1
List of Acronyms	1
Introduction.....	2
Gas composition and pressure measured during 3013 destructive examination	3
Gas composition at time of packaging.....	7
The amount of H ₂ and the H ₂ fraction	10
The H ₂ fraction for use in the 3013 pressure equation.....	17
Conclusions.....	17
Acknowledgements.....	18

Appendices

Appendix 1. Gas composition query.....	19
Appendix 2. The observed pressure and gas composition for 3013 containers that have undergone DE	25
Appendix 3. The volume of the inner container with convenience container, the material mass and particle density, the material volume, the free gas volume, the wattage of the material, and the best moisture percentage for each container.....	30
Appendix 4. The amount of each gas calculated from the pressure, the gas volume, and the measured percent of each gas (input data from Appendix 2 and Appendix 3) Reported percentages that were non-numeric (ND, Trace, and <0.01) are reported as --- in this table.....	35
Appendix 5. The calculated gas composition and the total pressure at the time the inner can was welded. In order to calculate the initial composition, 25% of the He generated during storage is subtracted from the amount of He observed at DE. The generated He typically represents a small fraction of the amount of He observed at DE.	40
Appendix 6. The amount of H ₂ observed, the amount of H ₂ calculated using the 3013 pressure equation, and the hydrogen fraction (ratio of observed to calculated).	45
Appendix 7. Estimating bounds for the H ₂ fraction in the 3013 container population from the DE observations.	49

Figures

Figure 1. Histograms of the Rocky Flats packaging pressures (red) and the Hanford packaging pressures (green). The dotted vertical lines are the average local atmospheric pressures with the same color code.....	9
Figure 2 The amount of H ₂ at DE as a function of the best wt% moisture for each packaging site.	10
Figure 3. The moles of H ₂ as a function of the moles of water on the packaged material at the time of packaging for each packaging site.....	12

Figure 4. The amount of H ₂ as a function of time for the packaging sites. There is no trend to lower amounts of H ₂ as packaging time increases.	13
Figure 5. The H ₂ fraction versus best wt% moisture for the packaging sites. The blue dashed line is the H ₂ fraction value from statistical analysis of the compliant population which excludes the HHMC point at 0.52 wt% moisture. The green dot-dash line is an empirical boundary that ties the H ₂ fraction statistical value of 0.13 used for the compliant population to the H ₂ fraction of 0.25 used for the entire population. Reported moisture is the best moisture.	15
Figure 6. The H ₂ fraction versus wt% best moisture with the small-scale reactor (SSR) and full-scale container (FSC) shelf-life data included to illustrate that the H ₂ fractions observed at DE are consistent with the H ₂ fractions observed in shelf-life data. The SSR H ₂ fractions are expected to be larger than the H ₂ fractions for either the 3013 containers or the FSCs (see text).	16

Tables

Table 1. The free inner container volume within the container configurations used by the packaging sites. ¹	5
Table 2. The elevation and average local atmospheric pressure of the packaging sites.	8
Table 3. The packaging pressures for Livermore and Savannah River.	8

Appendix 7 Figures

Appendix 7 Figure 1. The H ₂ fraction data for 114 observations. The small horizontal lines are the interval observations and the X's are the non-interval observations. The red curve is the beta cumulative distribution fitted to these data.	50
Appendix 7 Figure 2. Comparison of the beta-fit cumulative distribution based on all of the H ₂ Fraction data (red) to the empirical cumulative distribution (black).	51
Appendix 7 Figure 3. Shows the shape parameters for 10,000 resampling fits to the original data set.	52
Appendix 7 Figure 4. Histogram of bootstrap probabilities of H ₂ fraction less than 0.25.	53
Appendix 7 Figure 5. The H ₂ fraction data for 113 compliant observations. The small horizontal lines are the interval observations and the X's are the non-interval observations. The red curve is the beta cumulative distribution fitted to these data.	54
Appendix 7 Figure 6. Comparison of the beta-fit cumulative distribution based on the compliant H ₂ Fraction data (red) to the empirical cumulative distribution (black).	54

Appendix 7 Tables

Appendix 7 Table 1. Quantiles and upper 95% confidence limits for 95%, 99%, 99.6% and 99.9% probabilities.	52
Appendix 7 Table 2. The results of a beta distribution fit of the H ₂ fractions of the entire compliant population (HHMC excluded).	55

List of Acronyms

CH ₄	Methane
CO	Carbon Monoxide
CO ₂	Carbon Dioxide
CPD	Can Puncture Device
DE	Destructive Examination
DOE	Department of Energy
FSM	Field Surveillance Module
FY	Fiscal Year
GEST	Gas Evaluation Software Tool
H ₂	Hydrogen
He	Helium
HHMC	Hanford High Moisture Container
IC	Inner – Convenience
ID	Identification
ISMP	Integrated Surveillance and Monitoring Program
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
MIS	Material Identification and Surveillance
ND	Not Detected
N ₂	Nitrogen
N ₂ O	Nitrous Oxide
OI	Outer-Inner
O ₂	Oxygen
RFETS	Rocky Flats Environmental Technology Site
SPA	Surveillance Program Authority
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TGA-MS	Thermogravimetric Analysis – Mass Spectroscopy

Introduction

The DOE's 3013 Standard contains a methodology to calculate the maximum pressure within a 3013 container during storage.¹ This methodology is commonly referred to as the 3013 pressure equation. The 3013 pressure equation assumes all of the water within the sealed container undergoes decomposition to form hydrogen gas (H_2) with the oxygen component of the water forming a non-volatile chemical species. This assumption was thought to be conservative at the time the 3013 Standard was written. For instance, the inner container design required an indication, detectable by non-destructive means, of a buildup of internal pressure of 790 kPa (100 psig) or greater. In Appendix A of the 3013 Standard, which details the technical bases for the Standard's requirements, it is stated that "an internal pressure indication of 790 kPa (100 psig) is adequately indicative of *unexpected* pressurization, yet far below the design pressure for the outer container" (italics added by the authors).

There are a variety of reaction mechanisms that would lead to less than all of the available water decomposing to form H_2 . Reaction of water to form surface hydroxyls reduces the amount of water available to form H_2 .² Recombination reactions ($H_2 + \frac{1}{2} O_2 \rightarrow H_2O$) either in the gas phase initiated by radiation or on the plutonium dioxide surface, which is thought to be catalyzed, would result in a steady state between formation and recombination.³ Consumption reactions where H_2 reacts with components of the material to form non-volatile chemical species would reduce the H_2 pressure. Hydrated alkaline earth chlorides when mixed with PuO_2 have been shown to form HCl reducing the amount of hydrogen available to form H_2 .⁴ Ammonium chloride coating, which has also been observed in some DE containers, also reduces the hydrogen available to form H_2 as well as the N_2 present in the container.⁵ The ability to use rate constants associated with these reaction mechanisms to model the chemistry within sealed 3013 containers is rudimentary at best and the complexity of the packaged materials limits the usefulness of this approach at this time. In other words, although we know that not all of the available water will form H_2 , we do not know how to mechanistically estimate a reasonable value for the fraction of water that will form H_2 within a sealed 3013 container using known reactions.

One approach to estimate the maximum fraction of water that will form H_2 is to use empirical observations of H_2 gas composition in sealed containers versus time. This approach was used to determine that the maximum fraction of water that decomposed to form H_2 was 0.25 for Materials Identification and Surveillance (MIS) represented materials containing salts and loaded with nominally 0.5 wt% water in small-scale reactor studies.⁶ The small-scale reactor geometry is conservative with respect to 3013 containers for maximum H_2 pressures due to the reduced dose to the headspace gas.⁷ In the small-scale studies, the rate of H_2 pressure increase had slowed sufficiently for all materials that the H_2 pressure at 5 years had reached more than 95% of its projected maximum. For eight of the 11 materials studied, the H_2 pressure at 5 years was more than 99% of the projected maximum. In one case, the H_2 pressure decreased after reaching a maximum pressure, but the time for the H_2 partial pressure in this case to reach a non-detectable quantity would be decades.⁸

In this report, we use gas composition and pressure measurements taken during destructive examination (DE) of 3013 containers to estimate the maximum value of the fraction of water converted to H₂ in the entire population of 3013 containers, which we refer to as the H₂ fraction. All 3013 containers undergoing DE have been packaged for more than 5 years and should have reached their maximum H₂ pressure. There is no evidence for the hydrogen pressure to increase once it has stabilized or started to decrease in containers in a stable environment. First, we gather gas composition and gas pressure measured at DE, and relevant material properties from certified sources. Second, we vet the data to identify entries that need to be updated from primary source documents and develop a table of corrected data which documents the results. Third, we calculate the gas composition and pressure at packaging. Fourth, we calculate the H₂ fraction, the ratio of the moles of H₂ gas that was measured at DE to the moles of water that was present at packaging. Finally, we derive a description of the bounding H₂ fraction observed in the DE collection of containers. We argue that this bounding description is conservative because (1) the random selection of containers is meant to represent the whole population of containers, (2) the random selection was prioritized according to moisture with the highest moisture containers having been opened (mechanistically, higher moisture results in higher H₂ pressures and higher hydrogen fractions), and (3) the random sample is augmented with engineering judgement samples that were chosen mainly for their high moisture content.

All containers that have undergone DE were packaged when there was no RH requirement for salts. The containers packaged at RFETS, SRS and LLNL were packaged in dry atmospheres that meet the current 3013 Standard RH requirements for salts with RH less than 15%. In contrast, the containers packaged at Hanford were packaged in atmospheres where the RH varied as the outside RH varied and most if not all would not have met the current 3013 RH requirements for packaging salts. In the future, all salt bearing materials will be packaged in dry atmospheres.

Gas composition and pressure measured during 3013 destructive examination

The Department of Energy's Integrated Surveillance and Monitoring Program (ISMP) implements requirements to measure gas pressure and gas composition during DE of 3013 containers as required by DOE's 3013 Standard.⁹ There were one hundred and fourteen DEs conducted by Savannah River site personnel between 2007 and 2017. For each DE, the gas pressure and gas composition was determined. A brief description of the process is described with references to primary documents.

Two gas samples are collected and analyzed from each 3013 container undergoing DE. The gas samples are collected in K-Area and shipped to Savannah River National Laboratory for analysis. One gas sample is collected for the volume between the outer container and the inner container, referred to as the OI sample, and one gas sample is collected for the volume within the inner container, referred to as the IC sample. The IC sample is representative of the gas within the convenience container as well, because the convenience container is punctured at the same time. In addition, the convenience container is not sealed and the gas is expected to be the same as in the IC. Specially designed equipment at K-area known as the can puncture device (CPD) is

used to puncture the outer and inner containers and obtain the gas samples.¹⁰ The process for collecting samples is essentially the same for both samples. The process includes evacuating and backfilling with nitrogen three times to purge air from the CPD, evacuation of the CPD to less than 1 psia, a pre-puncture leak check, puncturing of the container (outer or inner/convenience), purging the lines leading to the sample cylinder to ensure gas from the volume of interest is collected, and sampling of the gas. Data is collected on appropriate forms and reviewed and certified by the Surveillance Program Authority (SPA). The whole process is controlled by Use Every Time written procedures which are detailed in the yearly processing plan for K-area.¹¹ An example, the processing plan for FY08, is given in Reference 8.

Pressure data are recorded during the entire process and are input to the final evaluation. Each gas sample is analyzed by calibrated micro gas chromatography at Savannah River National Laboratory and the results reported as volume fraction of the gas sample. Material particle density is measured on a sample of the oxide. A software package known as the Gas Evaluation Software Tool (GEST) was developed that is integrated with the operation of the CPD.¹² The GEST assumes ideal gas law behavior so that the initial pressure and composition within a container can be calculated from system pressures before and after puncture, known volumes, temperatures, and gas in-leakage rates and times. The GEST was benchmarked using statistically designed tests with various combinations of container pressures and compositions, CPD pressures, and container volumes.¹³ The GEST software is version controlled.¹⁴ A more detailed description of the process and uncertainties is available.¹⁵

The GEST calculates the pre-puncture 3013 gas fractions and pressure within the two volumes. For the inner container sample, GEST input includes the 3013 container configuration which determines the free volume within the inner container with the convenience container in place, the material net weight and particle density, the pressures measured before and after the container is punctured, the CPD leak rates and times, and the estimated average gas temperature for the CPD/3013 configuration. The GEST calculates a correction to the analyzed gas compositions to account for nitrogen remaining in the partially evacuated CPD prior to puncture and air in-leakage during the leak-testing, puncture, and gas sampling evolutions. The gases for which gas fractions are reported are He, H₂, N₂, O₂, CO₂, CO, N₂O, and CH₄.

The DE GEST results are entered into a Microsoft ACCESS database maintained by Savannah River National Laboratory. The ACCESS database contains all relevant information about stored materials and 3013 containers as required by the 3013 Standard and ISMP, and is the certified source for results of DE activities.¹ The database is classified, however, there is a controlled, unclassified subset of information primarily consisting of DE results from the ACCESS database called the Field Surveillance Module (FSM). The gas compositions and pressures reported here were mostly obtained from a query of the FSM. The FSM query is described in Appendix 1.

The data returned by the query were vetted using three approaches. First, the sum of all gas composition percentages was calculated and compared to 100%. In all cases, the sum was within rounding errors of 100%. Second, all of the returned values were inspected to ensure there was a valid entry. The format of the GEST reports changed over time resulting in some inconsistency in the values entered into the database. From 2007 to 2010, the gases below 0.1% were either not reported or reported as 0.000 in the database. In these cases, the values in the appropriate GEST

report are used. The data for FY15DE08 to FY17DE06 are not entered into the database as of the date of this report and the GEST reports for these DEs were used as the primary data source. In some cases, the number of significant figures varied. Percentages are reported here to 0.1%. In many cases, the uncertainty was not reported, especially for gas percentages less than 1%. For these cases, the uncertainty was taken from the GEST summary page of the appropriate GEST report.

For H002728 FY10DE10, an error in sampling occurred that introduced air into the sample. Two models were used in the GEST report to bound the gas percentages. The partial mixing model is reported here. In this model, the amount of oxygen is reported as -0.06%. The uncertainty for each gas used here was calculated as the difference between the models plus the uncertainties for each model.

For L000075 FY12DE05, helium was 2.9% and argon was 15.5%. In Appendix 2, the helium and argon content are combined and reported as helium.

The vetted gas pressures and gas component percentages are reported in Appendix 2. There are three terms used in the GEST reports and recorded in the database when the value is less than the official reporting range that describe a range of gas pressure rather than a value. The three terms are <0.10%, Trace, and ND. They are defined in Appendix 1 Table 2. In Appendix 2, if a gas percentage is not reported, then one of these terms is reported in the column for the gas percentage. There is no uncertainty associated with these terms.

In order to make comparisons between moles of gas produced by the various terms of the 3013 pressure equation and the gas pressure and percentages reported in Appendix 2, the free gas volume and average gas temperature need to be specified. The ideal gas equation can be used to calculate pressures from moles and moles from pressures when the volume and temperature are known. The free gas volume is calculated by GEST but is not recorded in the database. The free gas volume is calculated from the volume within the inner container, the material mass, and the measured material particle density. The volume within the inner container depends upon the container configuration. Various container configurations with their free volume are documented in the GEST calculation page. The free inner container volumes for the configurations used by the various packaging sites to date are given in Table 1. The free gas volume for each DE is calculated from the free inner can volume, the material mass, and the material density, Equation 1:

$$V_g = V_c - \frac{m}{\rho}$$

Equation 1

where V_g is the free gas volume, V_c is the free inner can volume for the packaging site, m is the mass of the material within the container, and ρ is the particle density of the material.

Table 1. The free inner container volume within the container configurations used by the packaging sites.¹

	Volume between	Error in Volume Between	Free Inner Can Volume	Error In Free Inner Can volume
--	----------------	-------------------------	-----------------------	--------------------------------

	Inner & Outer Cans	Inner and Outer Cans		
	L	L	L	L
RFETS	0.2710	0.0260	1.9780	0.0310
Bagless SRS	0.2930	0.0120	1.9940	0.0150
Bagless Hanford	0.2930	0.0120	1.9090	0.0220
LLNL	0.2710	0.0260	1.9780	0.0310
LANL	0.2350	0.0270	2.2490	0.0230

The average gas temperatures used by GEST are reported in the GEST reports but are not included in the database. It was found that the average gas temperature in the GEST reports was 155 °F for all DEs up to FY13 at which point the GEST calculation for determining the average gas temperature changed. The average gas temperature for DEs starting in FY13 to FY17DE06 were calculated based on the 3013 container heat generation and documented in the GEST reports.

An estimate of the water in the container at packaging is needed to calculate the H₂ fraction. We use the database entry “best moisture” for this calculation. The fraction of water is required to be measured at packaging with acceptance criterion of less than 0.5 wt% for the package to be compliant with the 3013 Standard. A number of measurement techniques were used – loss-on-ignition (LOI) to 1000 °C, thermogravimetric analysis (TGA) to 1000 °C, TGA with mass spectrometry detection of water (TGA-MS) to 1000 °C, and TGA with infrared spectroscopy detection of water (TGA-IR) to 1000 °C. LOI and TGA are conservative for material without uranium – the mass loss to 1000 °C contains species other than water, for instance salt content begins to evaporate resulting in mass loss at 800 °C. Packaging sites typically reported the most conservative value that meets the criterion. This value is entered into the database as the moisture value. This value is useful for ensuring 3013 Standard requirements are met, however, due to its conservative nature, it is not the most accurate information available as to the amount of moisture within packages. An effort was made to evaluate all moisture measurement data which resulted in a “best moisture” value being reported in the database.¹⁶ The biggest impact was for packages with TGA measurements only where TGA to 650 °C was used as the “best moisture”. For packages where LOI was used to determine the moisture, the LOI value is recorded as the “best moisture” because only the initial and final masses are available. In this document, the database entry for “best moisture” is used and whenever moisture is referred to it is the “best moisture”.

Information regarding the material from the database and GEST reports including material mass, material particle density, material wattage, average gas temperature within the inner container, and best moisture are reported in Appendix 3 along with calculated values for each container’s material volume and free gas volume. The various sites used different methods to measure moisture. Each container may have more than one moisture value reported. The best moisture is the value considered to be most accurate available.

The data in Appendix 2 and Appendix 3 are used to calculate the number of moles of the component gases, He, H₂, N₂, O₂, CO₂, CO, N₂O, and CH₄, within the inner container at the time

of DE using the ideal gas equation. These calculated values are reported in Appendix 4. When the reported percentages from Appendix 2 are either <0.10%, Trace or ND, an entry of --- is found for the corresponding value in Appendix 4. A crosscheck of the calculated total number of moles to the total number of moles reported in the GEST reports was conducted for 23 DEs. It was found that the calculated number of moles and the number of moles reported in the GEST reports were the same within the reported number of significant figures used in the calculation.

Gas composition at time of packaging

The gas composition and pressure at the time of packaging were not measured. However, these quantities are needed to quantify changes in the headspace gas within a package during storage. We attempt to estimate the gas composition and pressure at the time of packaging from the information available at DE. We make the following assumptions in our estimation process: (1) the trace gases, CO₂, CO, CH₄, N₂O, were not present initially, (2) the H₂ was not present initially, (3) the amount of N₂ initially is equal to the amount of N₂ plus the amount of N₂O at DE, i.e. N₂ was not consumed, (4) the initial O₂ was mostly consumed and the initial amount of O₂ can be estimated from the amount of N₂ initially using the O₂/N₂ ratio of air (except for the two LLNL containers and seven Hanford containers H001916, H001941, H001992, H002195, H002200, H002636, and H002750 in which the packaging glovebox atmosphere was N₂ and air was excluded), and (5) the He measured at DE is greater than the initial amount because some of the He generated during storage escapes the plutonium dioxide lattice and enters the gas phase. He gas was added to all of the 3013 containers at packaging to facilitate the He leak-check process and was the principal component in all of the containers except for the LLNL containers where He was a minor constituent. The assumption concerning N₂ ignores the formation of non-volatile NO_x species that accumulate on the surface of the material during storage. These species are detected during TGA-MS of the material after storage, but are generally a very small component of the gases detected. The loss of N₂ to form NH₄Cl, which has been observed as a film on the inner surface of the inner container, is also not accounted for.⁵ If a more precise estimate of the initial N₂ is desired, quantifying the NO_x species evolved during TGA-MS of DE samples is one approach that could be used. The possibility that the N₂ at DE is less than the N₂ initially affects the O₂ component proportionally. Together this effect should result in a lower estimate of the initial pressure than was actually present, but by how much is difficult to ascertain.

Helium is produced by alpha decay within the stored material. The amount of He from alpha decay that is released into the gas phase has been of interest to the fuel and heat source communities. Mulford has looked at Pu-238 heat source material aged over a range of 20 to 35 years and determined that the fraction of He from alpha decay that has escaped the solid over this range is approximately 0.26.¹⁷ The He generated over the storage time is calculated from the time of storage and the wattage using the conversion factor $6.232 \times 10^{-5} \text{ mol W}^{-1} \text{ yr}^{-1}$ from Equation 23 of the 3013 Standard.¹ This amount is reduced by the fraction entering the gas phase which is taken to be 0.25.

The calculated gas composition and pressure within the inner container at the time of packaging is reported in Appendix 5. The amount of He generated is reported along with the ratio of the moles of He generated to the moles of He observed at DE. Generally, this ratio is less than 0.1

and any errors in estimating the amount released to the gas phase will have only a minor effect. In the case of the LLNL containers, the He from alpha decay appears to represent a large fraction of the He in the containers at the time of DE. For FY12DE05 L000075 the ratio is 3.6 and for FY11DE12 L000178 the ratio is 1.233.

The pressure at packaging should be close to but slightly less than the local atmospheric pressure due to heating and leakage of the internal gas by the welding process at packaging. The elevation and average local atmospheric pressure for the elevation for each of the packaging sites is given in Table 2. There will be normal variations in the local atmospheric pressure of 5%.

Table 2. The elevation and average local atmospheric pressure of the packaging sites.

Site	Elevation (m)	Atmospheric pressure (kPa)
Hanford	122	99.9
Los Alamos	2225	77.3
Lawrence Livermore	195	99.0
Rocky Flats	1828	81.2
Savannah River	92	100.2

The estimated packaging pressures (from Appendix 5) for DE containers packaged at Livermore and Savannah River are shown in Table 3. The packaging pressures are all below the local atmospheric pressure for these two sites and have a range of 2.7 kPa except for the Savannah River container S001721 which is 4.7 kPa lower than the nearest other Savannah River container. All of the packaging pressures for Livermore and Savannah River are below the local atmospheric pressure.

Table 3. The packaging pressures for Livermore and Savannah River.

DE	3013 ID	Pressure (kPa)
FY11DE12	L000178	94.5
FY12DE05	L000075	91.9
FY10DE18	S001721	87.1
FY11DE02	S002129	93.0
FY11DE05	S001105	93.2
FY12DE10	S002250	93.2
FY14DE06	S002277	93.6
FY14DE07	S002116	91.8
FY15DE02	S002162	91.9

The number of containers packaged at Hanford and Rocky Flats that have undergone DE are substantially more than the number of containers packaged at Livermore and Savannah River that have undergone DE. In order to examine them, histograms of the Hanford containers and the

Rocky Flat containers are constructed. The two histograms show the number of containers as a function of estimated pressure at packaging, Figure 1. The range of packaging pressures is nearly 42 kPa for Hanford and 26 kPa for Rock Flats, which is considerably larger than the range of 7.4 kPa for Livermore and Savannah River. The distributions for the Hanford and Rocky Flats sites are surprisingly similar despite the fact that the local atmospheric pressure at Rocky Flats is nearly 20 kPa lower than Hanford's local atmospheric pressure. Almost all of the estimated packaging pressures of the Hanford containers are below the average local atmospheric pressure. In contrast, almost all of the packaging pressures for the Rocky Flats site are higher than the average local atmospheric pressure.

Different inner container designs may explain the different behaviors of the packaging pressure with respect to local atmospheric pressure of containers packaged at Rocky Flats and Hanford. The RFETS lid did not sit on a lip like the Hanford/SRS containers. The RFETS lid was a tight fit and was forced by a pneumatic piston down a tapered area of the wall and held in place by the piston during welding. This caused an interference metal-to-metal fit which would have had very little leakage during the final few mm of insertion and welding. In addition, the pneumatic piston drove the lid down an approximately 7" section of the inner container rapidly. Gas probably leaked around the lid during positioning but overall the process could have resulted in an internal

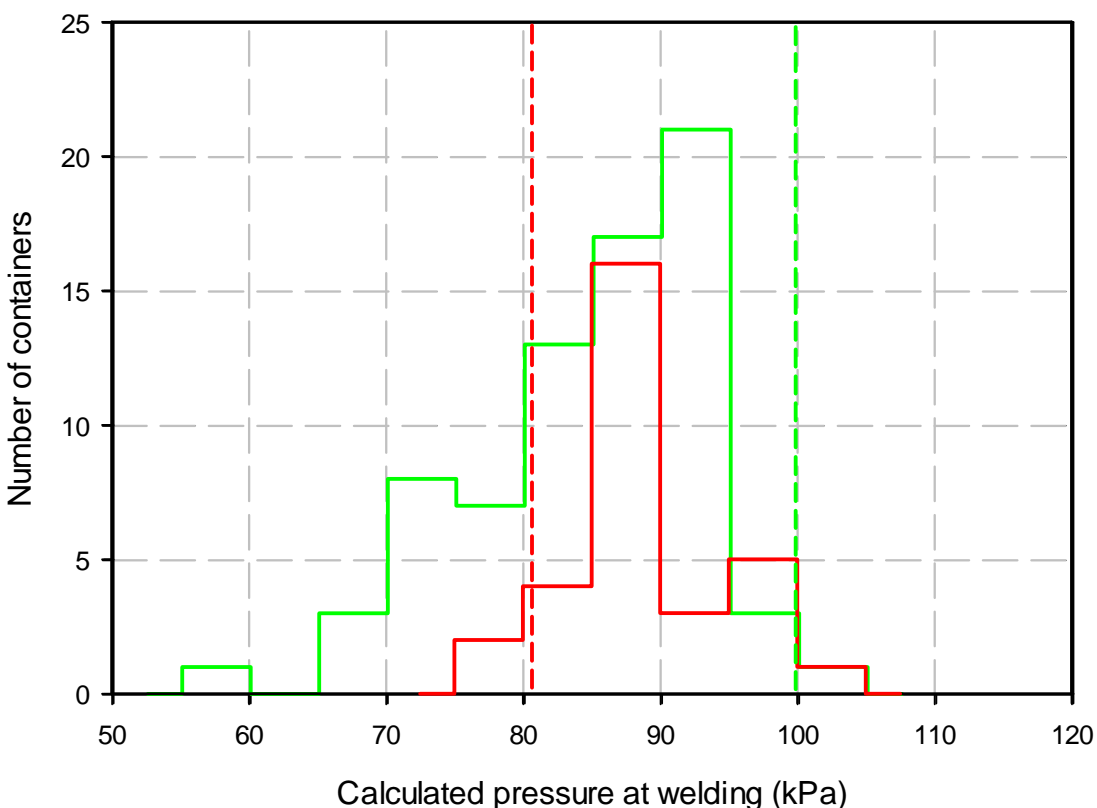


Figure 1. Histograms of the Rocky Flats packaging pressures (red) and the Hanford packaging pressures (green). The dotted vertical lines are the average local atmospheric pressures with the same color code.

pressure higher than local atmosphere after welding. The LLNL inner container and lid was the same design as RFETS, but the lid was inserted slowly with a manual screw mechanism which would have allowed sufficient time for gas to escape during positioning of the lid. In the Hanford/SRS design, the lid was a loose fit and sat on a lip during welding. This design allowed gas to leak around the lid resulting in approximately local atmospheric pressure in the container at the start of welding. The high temperatures associated with welding would have caused some expansion and leakage resulting in a pressure lower than local atmospheric after welding when the container has cooled.

The amount of H₂ and the H₂ fraction

The moles of H₂ in each container at DE have been calculated using the ideal gas equation with inputs for the total pressure, the hydrogen component of the total pressure, mass of material and particle density of the material, gas volume within an inner container minus the volume of the convenience container, and the average gas temperature. The results are documented in Appendix 5.

The moles of hydrogen are shown in Figure 2 as a function of wt% moisture in the material in each container as measured at the time of packaging. The graph shows an indication that for the

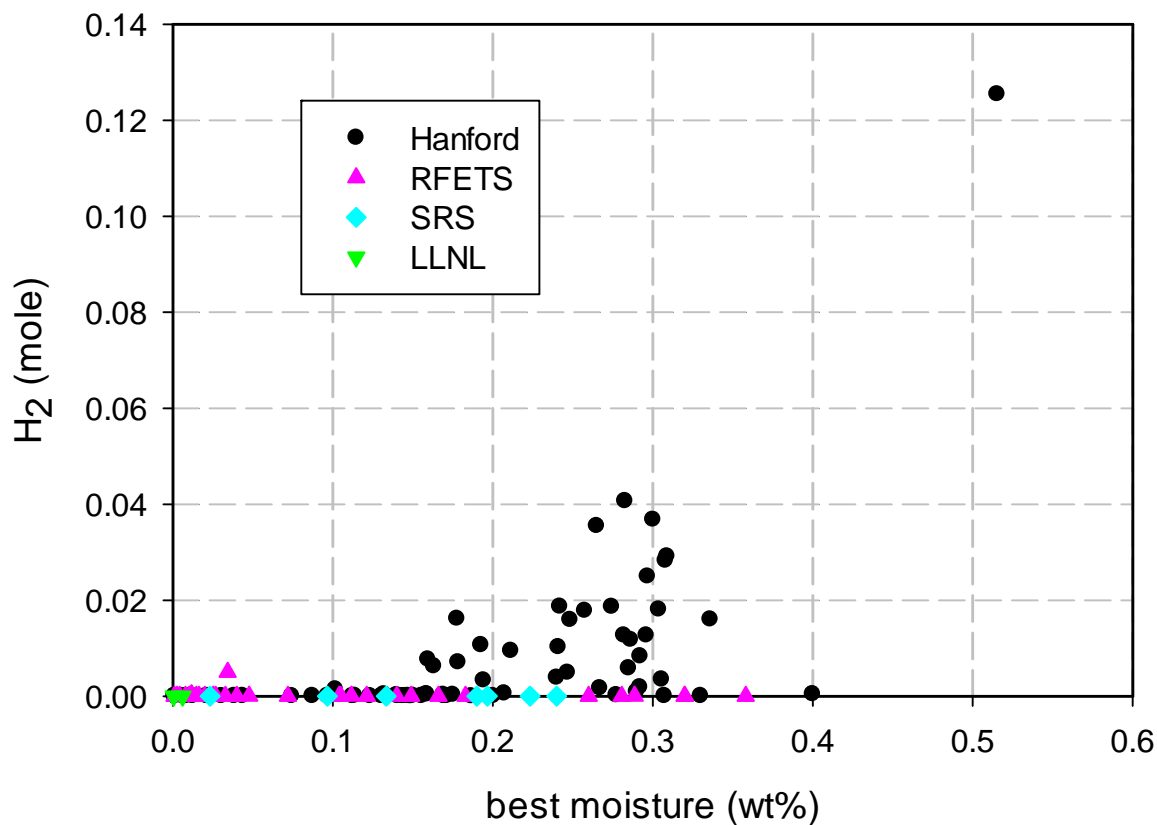


Figure 2 The amount of H₂ at DE as a function of the best wt% moisture for each packaging site.

Hanford subpopulation the extreme values of the absolute quantity of H_2 in a container increases with the wt% moisture of the material. The variability of the data is also increasing. Within the Hanford subpopulation, quantifiable H_2 (exceeding 0.1% of the total pressure) is only found in containers exceeding 0.1 wt% moisture. The Hanford High Moisture Container (HHMC) exceeds the 0.5 wt% moisture limit of DOE-STD-3013 and is therefore not a member of the 3013 certified population, but its moles of H_2 follows the extreme value trend established by the Hanford containers with less than 0.5 wt% moisture.

The SRS subpopulation has no containers with quantifiable H_2 . Five of the seven SRS containers fall in the >0.1 wt% moisture range where the Hanford subpopulation shows quantifiable H_2 . The RFETS subpopulation has two containers with quantifiable H_2 and both of these occur at low wt% moisture. R610960 FY12DE08 with 11.8% H_2 has 0.03 wt% moisture and R610785 FY10DE15 with 0.7% H_2 has 0.01 wt% moisture. There are seven RFETS containers that exceed 0.1 wt% moisture and none of these containers showed quantifiable H_2 . This behavior is opposite to the Hanford subpopulation trend which shows substantial H_2 in the gas phase only in containers that exceed 0.1 wt% moisture.

In the region above 0.24 wt% moisture, there are 24 Hanford containers. Of these containers, only four have less than 0.01 moles of H_2 . Most of the items in the Hanford subpopulation in this region contained over 2000 g of material, but these four containers each contained less than 900 g of material. Thus, despite high water mass fractions, the absolute amounts of water in these four containers are lower than the other 20 Hanford containers in this region. This suggests that plotting the H_2 as a function of the moles of water may provide some insight. Figure 3 shows the moles of H_2 as a function of the moles of water. In the region above 0.25 moles of water, there are 21 Hanford containers, 5 SRS containers, and 7 RFETS containers. One of the 21 Hanford containers has less than 0.01 moles of H_2 whereas all of the SRS and RFETS containers have less than 0.01 moles of H_2 . The lack of H_2 in the SRS and RFETS subpopulations is not consistent with expectations from shelf-life studies with moles of water proportional to the amount of material nor with the Hanford subpopulation.⁶ This could be due to moisture samples that do not represent the material or conservatism in the process for taking and handling samples for moisture measurement at SRS and RFETS that result in moisture uptake by the samples prior to measurement, i.e. the moisture samples may be conservative, but not necessarily an accurate representation of the packaged material. For RFETS and SRS, the stabilization, handling, and sampling were in a dried air box. The samples were then removed from the boxes and taken to another glovebox (not as dry) for analysis. For Hanford, the stabilization, handling, sampling, and analysis were all handled in a wet glovebox. This means that the RFETS and SRS oxides are more likely to have less actual moisture than the moisture sample compared with Hanford. This approach is good for ensuring the packaged material meets the 3013 criteria. However, if the moisture for these two sites is not accurate, then conclusions based on DE observations which include the amount of moisture as an argument could be suspect. The RFETS, SRS and LLNL subpopulations was packaged in dry atmospheres that meet the current 3013 Standard RH requirements for packaging salts whereas the Hanford subpopulation was packaged in an atmosphere that routinely exceeded 15% RH. The source of the difference between the Hanford subpopulation and the RFETS and SRS subpopulations should be explored further.

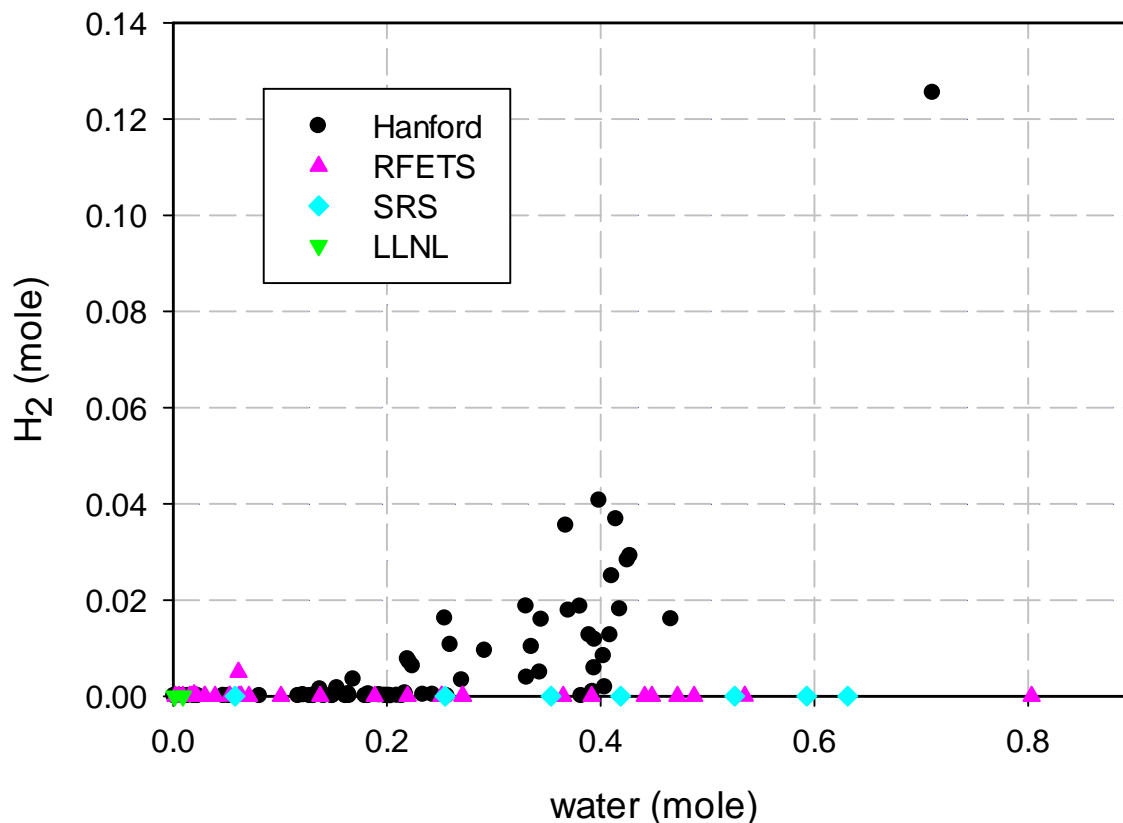


Figure 3. The moles of H₂ as a function of the moles of water on the packaged material at the time of packaging for each packaging site.

Determination of the amount of H₂ at DE is a single measurement in time for a system that changes with time. Those that are non-zero obviously showed a net increase during storage. Assuming a steady state has been reached between H₂ formation reactions and H₂ consumption reactions, any decrease in the rates of the H₂ generation reactions without a corresponding decrease in the rates of the H₂ consumption reactions will result in a new steady state at a lower H₂ partial pressure. In addition to ongoing generation mechanisms, there are mechanisms that can result in the H₂ partial pressure decreasing after reaching a maximum value. For instance, water can react with the material to form hydroxyls which do not seem to form H₂ thereby reducing the H₂ generation rate.⁸ The time for H₂ to reach a steady state, the time the system remains near that steady state, and the time for the H₂ partial pressure to decrease will vary with the system. In one shelf-life study, it took ~0.5 years to near a steady state pressure, it remained near that pressure for a year, and at 10 years the H₂ partial pressure had decreased but was still over 10% of the maximum value. On the other hand in another shelf-life study, the estimated maximum partial pressure of hydrogen was not reached until 5 years and has not decreased after 15 years.⁶ It would be useful to see if a reduction to lower amounts of H₂ in DE occurs as time during storage increases. Figure 4 shows no evidence of such a correlation, however.

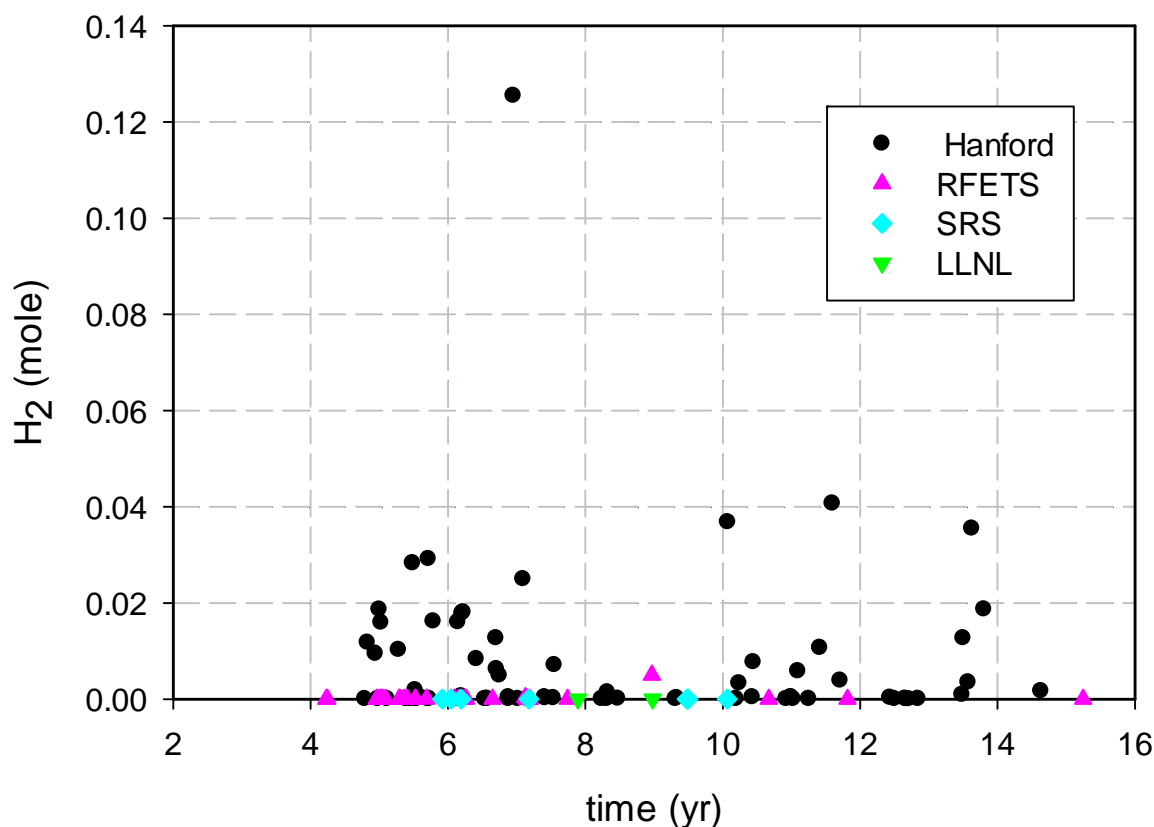


Figure 4. The amount of H₂ as a function of time for the packaging sites. There is no trend to lower amounts of H₂ as packaging time increases.

The 3013 Standard's pressure equation calculates the maximum H₂ pressure in 3013 containers by assuming that all of the water present undergoes decomposition to produce H₂ gas, all of which will remain in the gas phase and that the oxygen content of the decomposed water will be incorporated into the material. DE gas analyses allow us to look at how close actual results approach this limiting assumption. We define the H₂ conversion fraction or H₂ fraction as the mole ratio of H₂ measured at DE to water measured at packaging. Since each molecule of water can form one molecule of H₂, this is equivalent to the ratio of H₂ pressure measured at DE to the maximum H₂ pressure predicted by the Standard's pressure equation. The moles of water within the container is given by the product of the best moisture wt% times the material mass divided by the molecular weight of water. The relevant information to determine the moles of water is given in Appendix 3. The moles of H₂ are given in Appendix 4. The moles of H₂ gas at DE, the moles of H₂ predicted by the pressure equation (the same as the moles of water at packaging), and the H₂ fractions are given in Appendix 6. For this calculation, specific numerical values are adopted for the analytical results reported as <0.1%, Trace, and ND. In order to calculate the maximum H₂ fraction possible, the maximum value for each of these terms is used, i.e. 0.1% for <0.10%, 0.01% for Trace, and 0.001% for ND. This allows us to include all results in graphing and modeling the H₂ fraction even when no specific H₂ was reported due to calibration limitations. In some instances the value for the best moisture is negative or very small. A best

moisture value of 0.001% has been assumed for all reported moisture values less than 0.001% including negative reported values when calculating the H₂ fraction.

The resulting H₂ fractions vs the best wt% moisture for each DE are shown in Figure 5. The highest measured H₂ fraction to date is for the HHMC at just below 0.18. For the 3013 containers with less than 0.5 wt% moisture the maximum value is slightly greater than 0.10. The two RFETS containers in which there were measurable quantities of H₂ are more obviously unusual here than in the plots of the moles of H₂ because the wt% moisture is very low in these two RFETS containers. These two containers are anomalies in the SRS, RFETS and LLNL subpopulations.

The extreme values of the observed H₂ fraction appear to increase with moisture fraction, although there is considerable scatter. A beta distribution analysis of the data excluding the HHMC value at 0.52 wt% moisture results in a maximum H₂ fraction of 0.13 with 99% confidence that 97.5% of the population is less than this value (see Appendix 7 for details).

In Figure 5, the dashed blue line represents the 0.13 maximum H₂ fraction for the population excluding the HHMC and is plotted up to 0.31 wt% moisture. The density of data points begins to decline after ~0.31 wt% moisture. There is no H₂ fraction exceeding 0.1 in the interval between 0.31 and 0.4 wt% moisture. However, the largest value of ~0.18 occurs at 0.52 wt% moisture. It is likely that if there were more observations between 0.31 and 0.5 wt% moisture, then the trend in the extreme values of the H₂ fraction increasing with wt% moisture would continue in this region. The green dotted line in Figure 5 is an empirical boundary chosen to represent this potential trend in extreme values between 0.3 and 0.5 wt% moisture. One end of this line is anchored at 0.31 wt% moisture and 0.13 H₂ fraction. The other end is anchored at 0.25 H₂ fraction in the vicinity of 0.5 wt% moisture. A line with a slope of 0.60 (ΔH₂ fraction/Δwt%) meets these criteria with the slope having an appropriate number of significant figures. This empirical boundary line also takes into account expert judgement and the results of shelf-life studies. The expression for the boundary between 0.31 and 0.51 wt% moisture is given by

$$H_2 \text{ Fraction} = 0.6 \times \text{wt\% moisture} - 0.056$$

where the units of the slope are the same as the units of wt% moisture.

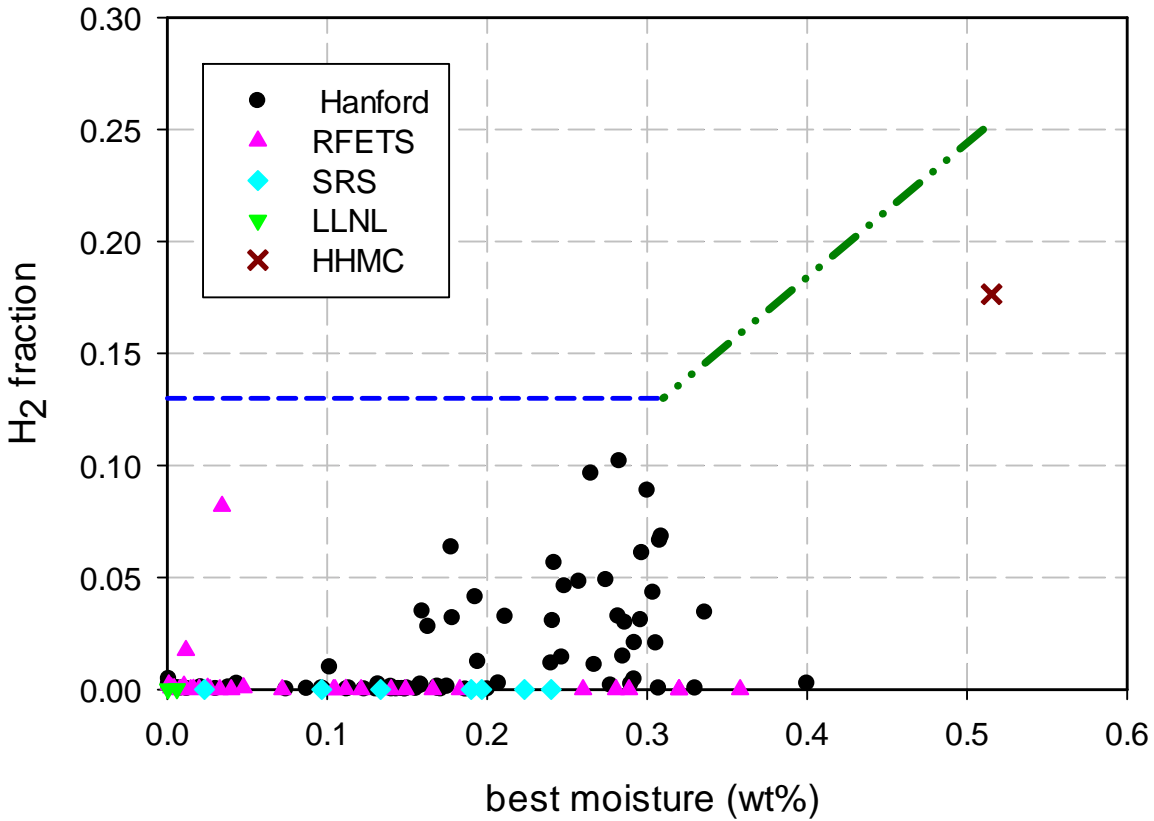


Figure 5. The H₂ fraction versus best wt% moisture for the packaging sites. The blue dashed line is the H₂ fraction value from statistical analysis of the compliant population which excludes the HHMC point at 0.52 wt% moisture. The green dot-dash line is an empirical boundary that ties the H₂ fraction statistical value of 0.13 used for the compliant population to the H₂ fraction of 0.25 used for the entire population. Reported moisture is the best moisture.

The observation that the H₂ fraction increases as the moisture content increases is not unique. The observations reported here are mainly from salt containing materials. In high-purity plutonium oxides, the efficiency of H₂ production per adsorbed dose, as described by the G-value for hydrogen – G(H₂), is seen to increase as the monolayer coverage increases which is another way of saying as the wt% moisture increases.¹⁸ For the observations in Figure 5, a constant G(H₂) would result in a constant H₂ fraction if the rate of H₂ consumption depended on the hydrogen pressure and not on the moisture. For instance, for a constant G(H₂), if the wt% moisture doubled, then the rate of H₂ produced would double and the pressure would have to double in order for the H₂ consumption to balance the H₂ production. The observation that as the wt% moisture increases the H₂ fraction increases suggests that G(H₂) increases with increases in wt% moisture as well. Scatter is taken to be reflective of differences inherent to each material.

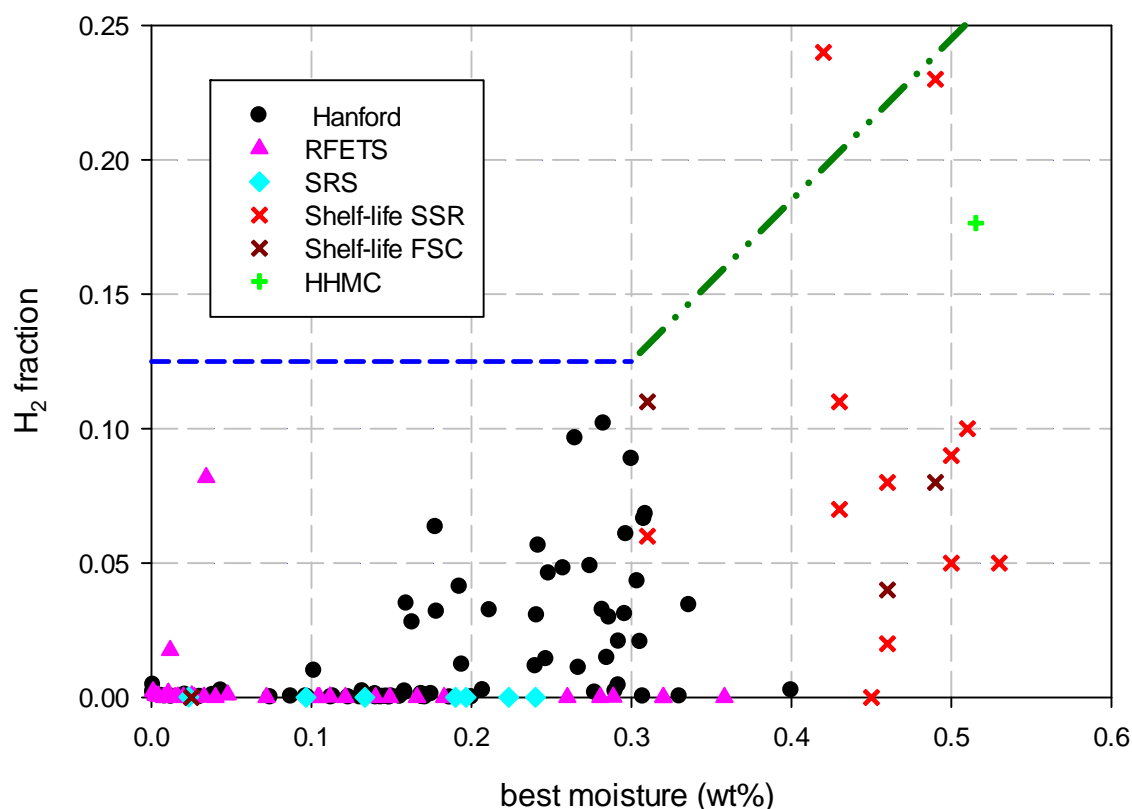


Figure 6. The H₂ fraction versus wt% best moisture with the small-scale reactor (SSR) and full-scale container (FSC) shelf-life data included to illustrate that the H₂ fractions observed at DE are consistent with the H₂ fractions observed in shelf-life data. The SSR H₂ fractions are expected to be larger than the H₂ fractions for either the 3013 containers or the FSCs (see text).

The H₂ fraction has been evaluated for represented materials in MIS shelf-life studies.⁶ The general class of materials that result in the largest pressure increases and the highest partial pressures of hydrogen in shelf-life studies are chloride salt bearing materials. The 3013 containers in Figure 5 with non-zero H₂ fractions contain chloride salt bearing materials. Figure 6 adds the shelf-life results to the plot shown in Figure 5. The shelf-life results are in agreement with the 3013 DE results in general, with the exception of two SSRs where the H₂ fraction is in the range of 0.21 to 0.25. The SSRs have less relative radiation dose to the headspace gas than either the 3013 containers or the shelf-life FSCs.^{7a} This reduction in relative dose to the headspace gas due to geometry is expected to reduce the magnitude of hydrogen consumption reactions compared to the larger containers, which in turn would result in higher H₂ fractions. Even in these two cases, the H₂ fraction is only 33% higher than the highest observed H₂ fraction in a 3013 container, which occurs at the highest wt% moisture. All the data available to the MIS program indicates that the H₂ fraction is 0.25 or less near 0.5 wt% moisture and decreases as the wt% moisture decreases. The SSRs are expected to have H₂ fractions larger than the 3013 containers. There are no shelf-life results that suggest the DE results are not bounding for

containers of that geometry. Therefore, it is reasonable to use the results of DE alone for determination of bounding values for the H₂ fraction.

The H₂ fraction for use in the 3013 pressure equation.

An expression for the maximum H₂ fraction for use in the 3013 pressure equation can be derived from the blue and green lines in Figure 5. There are two regions. From 0.0 to 0.31 wt% moisture, the H₂ fraction bound is based on statistical analysis of the data excluding the HHMC and is 0.13. From 0.31 to ~0.5 wt% moisture, the H₂ fraction is an empirical trend line given by $0.6 \text{ (units wt\% moisture}^{-1}) \times \text{the wt\% moisture} - 0.056$. Above 0.5 wt% moisture there is no data to determine the mathematical relationship. These expressions are believed to provide conservative values for the H₂ fraction. These expressions apply only to plutonium bearing materials within 3013 containers with measured moisture content between 0.0 and 0.5 wt%.

Conclusions

The gas composition and pressures of 3013 containers at DE have been extracted from the Field Surveillance Module database maintained by the ISMP and updated using GEST reports. The data have been vetted for accuracy. The H₂ partial pressure behavior for each packaging site has been examined. It is found that the SRS, RFETS and LLNL subpopulations behave differently than the Hanford subpopulation. We point out that the SRS, RFETS and LLNL subpopulations show essentially no H₂ gas at DE and they were packaged in dry atmospheres that meet the current requirements for chloride salt materials. The Hanford subpopulation shows a H₂ content that increases with amount of water with substantial variability of the data. For containers with similar amounts of material, the Hanford subpopulation tends to show an increase in the amount of H₂ with increase in wt% moisture with a threshold for observing quantifiable H₂ near 0.15 wt% moisture. However, at low material loadings the wt% moisture can be high with very low amounts of H₂ gas. Helium and nitrogen are the other major constituents. The oxygen partial pressure is essentially depleted at DE in all cases with the highest observed amount of oxygen being 1.7%, and in this case the H₂ concentration was essentially zero. Other minor gases observed in some cases included CO₂ with maximum content of 3.5%, CO with maximum content of 0.8%, N₂O with maximum content of 1.4%, and CH₄ with maximum content of 0.4%.

The ratio of H₂ measured at DE to the amount calculated using the 3013 pressure equation, referred to as the H₂ fraction, is calculated. In the DEs evaluated to date, the H₂ fraction varies from $\sim 1 \times 10^{-6}$ to 0.18. The maximum H₂ fraction value is 0.18 for the HHMC which has more than 0.5 wt% moisture. Based on this random sample of containers, it is very likely that the existing population of 3013 containers have H₂ fractions less than 0.25 according to statistical analysis and that below 0.31 wt% moisture it is likely that the H₂ fraction is less than 0.13.

These two bounding values have different statistical justifications. They are based not only on statistics, but on shelf-life studies and expert judgement. Therefore, it is recommended that for the purposes of calculating a maximum hydrogen pressure using the 3013 pressure equation, a correction to the amount of hydrogen produced from decomposition of the water be incorporated. The pressure (or moles) of H₂ should be reduced by multiplying by 0.13 for moisture content between 0.0 and 0.31 wt% moisture and by multiplying by the product of $0.6 \text{ (1/wt\% moisture)} \times$

Page | 17

wt% moisture minus 0.056 for moisture content between 0.31 wt% moisture and 0.5 wt% moisture. The appropriate moisture value for this calculation is the best moisture.

Acknowledgements

This work was supported by the Assistant Manager for Nuclear Material Stabilization in the Department of Energy's Office of Environmental Management, Savannah River Site. This work could not have been completed without the excellent work done by the technicians and staff who work in the glove boxes at K-Area and SRNL. The gas samples were collected at K-Area by Binh Nguyen's team consisting of Bill Biro, Darryl Ray, Roy Deiterich, Ron Bryant, Kristal Riley, Marc Cochran, Sam Cooper and John Kennedy. The gas samples were analyzed at Savannah River National Laboratory by Brenda Garcia-Diaz team (previously Kerry Dunn's team) consisting over time of Researchers: Ron Livingston, Ann Visser, Philip Almond, Glen Kessinger, Nick Bridges, Kim Crapse, Jon Duffey, and John Scogin and Technicians: Mona Blume, Beverly Wall, Betty Mealer, Dianne Scott, Minnie Hightower, Shirley McCollum, Mike Lee and Justin Doman. The ACCESS database and FSM are maintained by Buzzy Cheadle.

Appendix 1. Gas composition query

An SQL query was developed to return values from the database that are needed (1) to report the gas composition and pressure of containers when they underwent destructive evaluation and (2) to calculate the expected hydrogen pressure using the 3013 pressure equation. The values returned by the query and their use are described in Appendix 1 Table 1.

Appendix 1 Table 1. The information returned by the FSM query and what the information is used for in this report. When a value is used as an example it is indicated by (exp).

FSM Variable name	Unit or example	Use
3013ContainerID	H003328 (exp)	Identifies the specific 3013 container
SurveyFY	2007 (exp)	Year DE conducted
DENumber	7 (exp)	Number of DE that year; DE numbers are consecutive in the order the containers were opened.
GasSource	INNER-CONVENIENCE	Specifies that the gas information returned by the query is for the inner container.
Pressure	psia	Inner container pressure at time of DE.
UncertPressure	psia	Uncertainty in the pressure.
He	%	Percent of He within inner container at time of DE.
HeEOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertHe	%	Uncertainty in the He measurement.
H2	%	Percent of H ₂ within inner container at time of DE.
H2EOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertH2	%	Uncertainty in the H ₂ measurement.
N2	%	Percent of N ₂ within inner container at time of DE.

N2EOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertN2	%	Uncertainty in the N ₂ measurement.
O2	%	Percent of O ₂ within inner container at time of DE.
O2EOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertO2	%	Uncertainty in the O ₂ measurement.
CO2	%	Percent of CO ₂ within inner container at time of DE.
CO2EOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
Uncert CO2	%	Uncertainty in the CO ₂ measurement.
CO	%	Percent of CO within inner container at time of DE.
COEOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertCO	%	Uncertainty in the CO measurement.
N2O	%	Percent of N ₂ O within inner container at time of DE.
N2OEOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertN2O	%	Uncertainty in the N ₂ O measurement.
CH4	%	Percent of CH ₄ within inner container at time of DE.
CH4EOR	<0.10, trace, ND (exp)	Used to record estimated amounts of gases that were too low to quantify.
UncertCH4	%	Uncertainty in the CH ₄ measurement.
MCA3013NetMaterialWeight	g	Used to calculate the displaced volume of the material.
Wattage	W	Used to calculate the amount of He generated during storage.

AvgDensity	g mL ⁻¹	Used to calculate the displaced volume of the material.
InnerCanDateTimeofweld	10/1/2002 (exp)	Used to calculate the storage duration.
InnerConveniencePunctureDate	10/1/2002 (exp)	Used to calculate the storage duration.
BestMoisture	%	Used to calculate the amount of water in the container at packaging

The three terms <0.10%, Trace, and ND are used when the gas percentage for the species is less than the percentage range over which the micro GC is calibrated. These terms are recorded under the xEOR variable name where x is one of the eight gases that are reported by GEST, e.g. HeEOR. If one of these terms are reported under the xEOR variable name, then there is no entry under the x variable name. The definition of these three terms are given in Appendix 1 Table 2

Appendix 1 Table 2. The three terms used under the xEOR variable name in the Access database and their definitions.

EOR term	Definition
<0.10%	Peak is visually present and integrates greater than 0.01 vol. %, but less than 0.1 vol. %.
Trace	Visually a peak is greater than 3x the background noise and integrates to 0.01 vol. % or less. Note: a range of 0.01 vol % to 0.001vol % is used for this term in the evaluation of the maximum H ₂ fraction.
ND	No peak or if a peak is present and is visually less than 3x the background noise. Note: a range of 0.001vol % to 0 vol. % is used for this term in the evaluation of the maximum H ₂ fraction.

Description of query named “dkv July 13 2017 Gas Composition”:

1. The Query statement is:

```
SELECT DISTINCT SRNLCalcGas.[3013ContainerID], SurveyDocumentNumber.SurveyFY, SurveyDocumentNumber.DENumber,
SRNLCalcGas.GasSource, SRNLCalcGas.Pressure, SRNLCalcGas.UncertPressure, SRNLCalcGas.He, SRNLCalcGas.HeEOR,
```

SRNLCalcGas.UncertHe, SRNLCalcGas.H2, SRNLCalcGas.H2EOR, SRNLCalcGas.UncertH2, SRNLCalcGas.N2, SRNLCalcGas.N2EOR, SRNLCalcGas.UncertN2, SRNLCalcGas.O2, SRNLCalcGas.O2EOR, SRNLCalcGas.UncertO2, SRNLCalcGas.CO2, SRNLCalcGas.CO2EOR, SRNLCalcGas.UncertCO2, SRNLCalcGas.CO, SRNLCalcGas.COEOR, SRNLCalcGas.UncertCO, SRNLCalcGas.N2O, SRNLCalcGas.N2OEOR, SRNLCalcGas.UncertN2O, SRNLCalcGas.CH4, SRNLCalcGas.CH4EOR, SRNLCalcGas.UncertCH4, DE3013Surveillance.MCA3013NetMaterialWeight, tblPCDCalGamma.Wattage, SRNLSolidOxideAnalysis.AvgDensity, tblPCDInnerCan.InnerCanDateTimeofweld, DEGasSample.InnerConveniencePunctureDate

FROM (((((SRNLCalcGas INNER JOIN DE3013Surveillance ON SRNLCalcGas.[3013ContainerID] = DE3013Surveillance.[3013ContainerID]) INNER JOIN tblPCDCalGamma ON DE3013Surveillance.[3013ContainerID] = tblPCDCalGamma.[3013ContainerID]) INNER JOIN tblPCDInnerCan ON tblPCDCalGamma.[3013ContainerID] = tblPCDInnerCan.[3013ContainerID]) INNER JOIN DEGasSample ON tblPCDInnerCan.[3013ContainerID] = DEGasSample.[3013ContainerID]) INNER JOIN SurveyDocumentNumber ON DEGasSample.[3013ContainerID] = SurveyDocumentNumber.[3013ContainerID]) INNER JOIN SRNLSolidOxideAnalysis ON SurveyDocumentNumber.[3013ContainerID] = SRNLSolidOxideAnalysis.[3013ContainerID]

WHERE (((SurveyDocumentNumber.DENumber) Is Not Null) AND ((SRNLCalcGas.GasSource)="INNER-CONVENIENCE") AND ((SRNLSolidOxideAnalysis.AvgDensity) Is Not Null));

Appendix 1 Table 3. Structure of the Query in Design View, the 3013ContainerID linked the input tables.

TABLE	FIELD	CRITERIA
SRNLCalGas	3013ContainerID	
SurveyDocumentNumber	SurveyFY	
	DENumber	Is Not Null
SRNLCalGas	GasSource	“INNER-CONVENIENCE
	Pressure	
	UncertPressure	
	He	
	HeEOR	
	UncertHe	
	H2	
	H2EOR	
	UncertH2	
	N2	
	N2EOR	
	UncertN2	
	O2	
	O2EOR	
	UncertO2	
	CO2	
	CO2EOR	
	Uncert CO2	
	CO	
	COEOR	
	UncertCO	
	N2O	
	N2OEOR	
	UncertN2O	
	CH4	
	CH4EOR	
	UncertCH4	

DE3013Surveillance	MCA3013NetMaterialWeight	
tblPCDCalGamma	Wattage	
SRNLSolidOxideAnalysis	AvgDensity	Is Not Null
tblPCDInnerCan	InnerCanDateTimeofweld	
DEGasSample	InnerConveniencePunctureDate	

Appendix 2. The observed pressure and gas composition for 3013 containers that have undergone DE

3013 Container	FY	DE	Pressure (psia)	He (%)	H ₂ (%)	N ₂ (%)	O ₂ (%)	CO ₂ (%)	CO (%)	N ₂ O (%)	CH ₄ (%)
R600885	2007	1	12.8 +/- 0.4	90.8 +/- 2.8	Trace	9.2 +/- 1.0	<0.10	Trace	ND	Trace	ND
R601722	2007	2	12.5 +/- 0.4	87.2 +/- 2.8	Trace	12.7 +/- 1.1	<0.10	ND	ND	Trace	ND
R601957	2007	3	12.8 +/- 0.4	89.6 +/- 2.8	Trace	10.4 +/- 1.0	ND	Trace	ND	Trace	ND
R600719	2007	4	12.9 +/- 0.5	83.8 +/- 2.7	Trace	16.2 +/- 1.1	ND	Trace	ND	Trace	ND
R610735	2007	5	12.4 +/- 0.5	82.3 +/- 2.7	Trace	17.5 +/- 1.1	ND	0.2 +/- 0.01	ND	ND	ND
R610697	2007	6	12.5 +/- 0.5	82.0 +/- 2.7	Trace	18.0 +/- 1.1	ND	Trace	ND	ND	Trace
R601285	2007	7	13.4 +/- 0.5	77.4 +/- 2.6	Trace	22.5 +/- 1.2	ND	Trace	ND	ND	<0.1
R602731	2008	1	12.3 +/- 0.4	83.9 +/- 2.7	Trace	16.1 +/- 1.1	<0.10	Trace	ND	ND	Trace
R601318	2008	2	12.6 +/- 0.4	91.5 +/- 2.9	Trace	8.5 +/- 1.0	ND	Trace	ND	ND	ND
H000898	2008	3	12.3 +/- 0.4	80.3 +/- 2.7	Trace	19.3 +/- 1.2	<0.10	0.3 +/- 0.02	ND	ND	ND
R610327	2008	4	12.4 +/- 0.5	82.8 +/- 2.7	Trace	16.7 +/- 1.2	ND	0.4 +/- 0.02	ND	ND	ND
R610298	2008	5	13.0 +/- 0.5	84.0 +/- 2.7	Trace	16.0 +/- 1.1	ND	Trace	ND	ND	ND
R610324	2008	6	13.1 +/- 0.5	82.3 +/- 2.7	Trace	17.7 +/- 1.2	ND	ND	ND	ND	ND
H001992	2008	7	13.3 +/- 0.4	57.8 +/- 2.4	Trace	42.2 +/- 1.6	ND	ND	ND	ND	ND
H003157	2008	8	12.4 +/- 0.4	54.3 +/- 2.4	Trace	44.0 +/- 1.7	1.7 +/- 0.2	Trace	ND	ND	ND
R610584	2008	9	13.7 +/- 0.5	84.6 +/- 2.8	Trace	14.8 +/- 1.1	ND	0.7 +/- 0.03	ND	ND	ND
R610578	2008	10	12.5 +/- 0.5	82.1 +/- 2.7	Trace	17.8 +/- 1.2	ND	<0.10	ND	ND	ND
H001916	2008	11	13.3 +/- 0.4	54.5 +/- 2.4	Trace	45.6 +/- 1.8	ND	Trace	ND	ND	ND
H002088	2008	12	12.4 +/- 0.4	58.4 +/- 2.4	Trace	41.6 +/- 1.6	ND	Trace	ND	ND	ND
H003409	2008	13	13.4 +/- 0.4	47.1 +/- 2.3	18.7 +/- 0.3	34.2 +/- 1.5	ND	Trace	ND	ND	Trace
H002573	2008	14	17.6 +/- 0.4	37.9 +/- 2.2	27.9 +/- 0.5	33.9 +/- 1.4	ND	0.3 +/- 0.02	ND	<0.10	Trace
H002534	2008	15	13.7 +/- 0.4	41.4 +/- 2.3	30.8 +/- 0.5	24.0 +/- 1.4	ND	2.7 +/- 0.1	0.8 +/- 0.04	Trace	0.4 +/- 0.02
R610679	2008	16	11.7 +/- 0.4	70.3 +/- 2.6	Trace	26.7 +/- 1.4	ND	3.0 +/- 0.2	<0.1%	Trace	ND

3013 Container	FY	DE	Pressure (psia)	He (%)	H ₂ (%)	N ₂ (%)	O ₂ (%)	CO ₂ (%)	CO (%)	N ₂ O (%)	CH ₄ (%)
H002750	2008	17	13.1 +/- 0.4	63.1 +/- 2.5	Trace	36.8 +/- 1.8	0.1 +/- 0.3	Trace	ND	<0.10	ND
H004099	2009	1	11.7 +/- 0.4	45.0 +/- 2.3	Trace	53.0 +/- 2.0	0.2 +/- 0.2	0.4 +/- 0.1	Trace	1.4 +/- 0.1	ND
H004111	2009	2	14.4 +/- 0.4	34.1 +/- 2.2	21.5 +/- 0.4	44.4 +/- 1.8	<0.10	Trace	ND	<0.10	ND
H002554	2009	3	12.4 +/- 0.4	45.3 +/- 2.3	22.1 +/- 0.3	32.6 +/- 1.4	Trace	Trace	ND	Trace	Trace
H001941	2009	4	13.2 +/- 0.4	50.2 +/- 2.3	Trace	49.8 +/- 1.7	ND	Trace	ND	<0.10	ND
R602498	2009	5	12.3 +/- 0.4	87.5 +/- 2.8	ND	12.5 +/- 1.0	<0.10	Trace	ND	<0.10	ND
H002509	2009	6	20.6 +/- 0.5	34.1 +/- 2.2	36.3 +/- 0.6	29.2 +/- 1.4	<0.10	0.4 +/- 0.02	ND	<0.10	ND
H002565	2009	7	11.1 +/- 0.4	55.8 +/- 2.4	4.4 +/- 0.1	39.8 +/- 1.5	<0.10	<0.10	ND	<0.10	Trace
H002657	2009	8	11.2 +/- 0.4	61.8 +/- 2.5	0.7 +/- 0.1	37.5 +/- 1.5	<0.10	ND	ND	<0.10	ND
R611398	2009	9	11.4 +/- 0.4	80.6 +/- 2.7	<0.10	18.9 +/- 1.2	<0.10	ND	ND	0.4 +/- 0.02	ND
H002200	2009	10	13.5 +/- 0.4	50.5 +/- 2.3	<0.10	49.5 +/- 1.8	<0.10	ND	ND	<0.10	ND
H002667	2009	11	11.6 +/- 0.4	53.7 +/- 2.4	<0.10	46.2 +/- 1.7	<0.10	Trace	ND	Trace	ND
H002715	2009	12	20.5 +/- 0.5	34.4 +/- 2.2	38.5 +/- 0.6	24.2 +/- 1.2	<0.10	2.6 +/- 0.1	0.2 +/- 0.1	Trace	0.1 +/- 0.01
R610700	2009	13	11.5 +/- 0.4	82.9 +/- 2.7	<0.10	14.2 +/- 1.1	<0.10	2.9 +/- 0.2	ND	ND	ND
R610764	2009	14	12.2 +/- 0.4	83.3 +/- 2.7	<0.10	16.8 +/- 1.1	<0.10	ND	ND	ND	ND
R610573	2009	15	13.7 +/- 0.5	85.5 +/- 2.8	Trace	14.2 +/- 1.0	<0.10	ND	ND	0.3 +/- 0.01	ND
R610558	2009	16	12.3 +/- 0.4	81.8 +/- 2.7	Trace	17.2 +/- 1.1	0.1 +/- 0.2	0.3 +/- 0.1	ND	0.6 +/- 0.1	ND
R610806	2009	17	12.2 +/- 0.5	84.3 +/- 2.7	<0.10	15.1 +/- 1.1	<0.10	ND	<0.10	0.6 +/- 0.03	ND
H003119	2009	18	11.7 +/- 0.4	56.6 +/- 2.4	<0.10	43.3 +/- 1.6	<0.10	ND	ND	0.2 +/- 0.01	ND
H002195	2009	19	13.7 +/- 0.4	45.6 +/- 2.3	Trace	54.4 +/- 1.9	<0.10	<0.10 +/- 0	<0.10	ND	ND
H004251	2010	1	17.5 +/- 0.5	53.4 +/- 2.4	27.4 +/- 0.4	16.2 +/- 1.1	<0.10	2.5 +/- 0.1	0.4 +/- 0.02	<0.10	0.2 +/- 0.01
H002496	2010	2	11.6 +/- 0.4	54.3 +/- 2.4	1.3 +/- 0.1	44.4 +/- 1.7	<0.10	ND	ND	<0.10	ND
H003710	2010	3	16.6 +/- 0.4	39.8 +/- 2.2	25.7 +/- 0.4	34.0 +/- 1.4	<0.10	0.4 +/- 0.02	<0.10	<0.10	<0.10
H003655	2010	4	17.6 +/- 0.5	43.0 +/- 2.3	27.4 +/- 0.4	29.3 +/- 1.3	<0.10	0.3 +/- 0.01	<0.10	<0.10	<0.10
H002447	2010	5	12.0 +/- 0.4	52.0 +/- 2.4	<0.10	47.6 +/- 1.8	0.1 +/- 0.2	ND	<0.10	0.3 +/- 0.02	ND
R610627	2010	6	14.7 +/- 0.6	85.1 +/- 2.8	Trace	14.8 +/- 1.2	<0.10	<0.10	<0.10	ND	ND
H003900	2010	7	17.0 +/- 0.5	44.1 +/- 2.3	30.1 +/- 0.5	25.5 +/- 1.2	<0.10	0.2 +/- 0.01	ND	ND	<0.10

3013 Container	FY	DE	Pressure (psia)	He (%)	H ₂ (%)	N ₂ (%)	O ₂ (%)	CO ₂ (%)	CO (%)	N ₂ O (%)	CH ₄ (%)
H003650	2010	8	12.8 +/- 0.4	52.2 +/- 2.3	16.9 +/- 0.3	30.9 +/- 1.3	<0.10	ND	ND	<0.10	ND
H002567	2010	9	11.7 +/- 0.4	57.9 +/- 2.4	<0.10	42.1 +/- 1.6	<0.10	ND	ND	<0.10	ND
H002728	2010	10	11.4 +/- 0.4	54.0 +/- 7.6	14.7 +/- 1.7	31.3 +/- 5.8	-0.1 +/- 0.2	<0.10	ND	<0.10	ND
H002786	2010	11	12.5 +/- 0.4	51.7 +/- 2.3	10.5 +/- 0.2	37.7 +/- 1.5	<0.10	<0.10	<0.10	<0.10	ND
H003077	2010	12	12.2 +/- 0.4	52.3 +/- 2.4	<0.10	46.7 +/- 1.7	<0.10	ND	ND	1.0 +/- 0.05	ND
H003367	2010	13	11.5 +/- 0.4	61.2 +/- 2.5	0.8 +/- 0.1	38.0 +/- 1.5	<0.10	<0.10	<0.10	<0.10	ND
H003704	2010	14	15.3 +/- 0.4	41.3 +/- 2.2	22.5 +/- 0.4	36.0 +/- 1.5	<0.10	<0.10	<0.10	<0.10	<0.10
R610785	2010	15	13.9 +/- 0.5	84.8 +/- 2.7	0.7 +/- 0.1	14.5 +/- 1.1	<0.10	Trace	<0.10	ND	ND
R610826	2010	16	12.0 +/- 0.5	88.5 +/- 2.8	ND	11.5 +/- 1.1	<0.10	<0.10	ND	ND	ND
R610853	2010	17	12.3 +/- 0.5	86.8 +/- 2.8	Trace	13.2 +/- 1.1	<0.10	Trace	ND	ND	ND
S001721	2010	18	12.0 +/- 0.4	73.6 +/- 2.6	Trace	26.2 +/- 1.3	<0.10	Trace	ND	0.1 +/- 0.01	ND
H003443	2011	1	18.1 +/- 0.5	35.7 +/- 2.2	36.6	27.6 +/- 1.2	Trace	<0.10	ND	<0.10	ND
S002129	2011	2	12.4 +/- 0.4	59.9 +/- 2.4	Trace	39.7 +/- 1.6	Trace	Trace	ND	0.4 +/- 0.02	ND
H002592	2011	3	11.8 +/- 0.4	60.0 +/- 2.4	Trace	39.9 +/- 1.6	Trace	ND	ND	<0.10	ND
H003337	2011	4	12.3 +/- 0.4	62.7 +/- 2.5	Trace	37.2 +/- 1.5	<0.10	Trace	ND	Trace	ND
S001105	2011	5	12.0 +/- 0.4	45.2 +/- 2.3	trace	53.9 +/- 1.9	ND	Trace	ND	0.9 +/- 0.05	ND
H003343	2011	6	11.6 +/- 0.4	55.3 +/- 2.4	Trace	44.2 +/- 1.6	<0.10	ND	ND	0.5 +/- 0.03	ND
H003371	2011	7	12.1 +/- 0.4	56.7 +/- 2.4	15.1 +/- 0.2	28.2 +/- 1.3	ND	Trace	ND	<0.10	ND
H003526	2011	8	10.0 +/- 0.4	65.5 +/- 2.5	0.5 +/- 0.1	34.0 +/- 1.5	Trace	Trace	ND	Trace	ND
H003565	2011	9	10.5 +/- 0.4	67.9 +/- 2.5	0.9 +/- 0.1	31.1 +/- 1.4	<0.10	Trace	ND	Trace	ND
R611131	2011	10	13.8 +/- 0.5	86.1 +/- 2.8	ND	13.9 +/- 1.1	Trace	Trace	ND	ND	ND
H003625	2011	11	11.8 +/- 0.4	78.2 +/- 2.7	0.5 +/- 0.1	21.3 +/- 1.2	<0.10	ND	ND	ND	ND
L000178	2011	12	13.8 +/- 0.5	2.3 +/- 2.1	Trace	97.7 +/- 3.0	<0.10	Trace	ND	Trace	ND
H003328	2011	13	43.4	14.2	74.4	11.3	0.1	ND	ND	ND	ND
H001209	2012	1	12.1 +/- 0.4	55.0 +/- 2.4	0.4 +/- 0.1	44.6 +/- 1.6	Trace	ND	ND	ND	ND
H002574	2012	2	12.8 +/- 0.4	91.8 +/- 2.8	ND	8.1 +/- 1.0	Trace	ND	ND	<0.10	ND
H001513	2012	3	11.3 +/- 0.4	52.8 +/- 2.3	<0.10	46.5 +/- 1.7	<0.10	ND	ND	0.5 +/- 0.03	ND

3013 Container	FY	DE	Pressure (psia)	He (%)	H ₂ (%)	N ₂ (%)	O ₂ (%)	CO ₂ (%)	CO (%)	N ₂ O (%)	CH ₄ (%)
H003390	2012	4	11.4 +/- 0.4	57.7 +/- 2.4	0.2 +/- 0.1	41.9 +/- 1.6	<0.10	ND	ND	0.2 +/- 0.01	ND
L000075	2012	5	14.2 +/- 0.5	18.4 +/- 2.1	Trace	77.2 +/- 2.5	<0.10	3.5 +/- 0.2	<0.10	0.9 +/- 0.05	ND
H004012	2012	6	10.9 +/- 0.4	48.4 +/- 2.3	<0.10	51.5 +/- 1.8	<0.10	<0.10	ND	Trace	ND
H004048	2012	7	9.8 +/- 0.4	51.9 +/- 2.3	4.1 +/- 0.1	43.5 +/- 1.6	<0.10	0.4 +/- 0.02	<0.10	Trace	<0.10
R610960	2012	8	13.0 +/- 0.5	87.4 +/- 2.8	11.7 +/- 0.2	0.8 +/- 1.0	Trace	Trace	ND	ND	<0.10
S002250	2012	10	13.6 +/- 0.4	73.2 +/- 2.6	Trace	21.5 +/- 1.2	1.0 +/- 0.2	3.6 +/- 0.2	0.3 +/- 0.02	0.4 +/- 0.02	ND
H001236	2013	1	12.4 +/- 0.4	50.5 +/- 3.0	0.7 +/- 0.1	48.8 +/- 1.7	ND	ND	ND	Trace	ND
R610996	2014	1	12.2 +/- 0.5	92.5 +/- 4.4	<0.10	7.4 +/- 1.0	ND	Trace	ND	ND	ND
H003064	2014	2	22.9 +/- 0.5	45.6 +/- 2.8	43.9 +/- 1.9	10.2 +/- 1.0	ND	0.2 +/- 0.01	Trace	Trace	Trace
H003307	2014	3	11.2 +/- 0.4	54.6 +/- 3.1	<0.10	45.4 +/- 1.7	ND	Trace	ND	Trace	ND
H003052	2014	4	10.4 +/- 0.4	65.3 +/- 3.4	7.9 +/- 0.4	26.8 +/- 1.3	ND	Trace	ND	Trace	ND
H003898	2014	5	11.7 +/- 0.4	53.7 +/- 3.1	16.7 +/- 0.7	29.5 +/- 1.3	ND	0.1 +/- 0.01	ND	ND	Trace
S002277	2014	6	12.2 +/- 0.4	46.4 +/- 3.1	Trace	53.4 +/- 1.3	ND	ND	ND	0.2 +/- 0.01	ND
S002116	2014	7	13.0 +/- 0.4	80.2 +/- 4.0	ND	19.8 +/- 1.1	ND	ND	ND	Trace	ND
H004219	2014	8	10.5 +/- 0.4	68.9 +/- 3.6	0.9 +/- 0.1	30.2 +/- 1.3	ND	Trace	ND	Trace	ND
H002636	2014	9	12.3 +/- 0.4	50.9 +/- 3.0	Trace	49.0 +/- 1.7	ND	ND	ND	<0.01	ND
R610156	2015	1	12.5 +/- 0.4	83.2 +/- 4.1	ND	16.8 +/- 1.1	ND	ND	ND	ND	ND
S002162	2015	2	12.1 +/- 0.4	49.9 +/- 2.9	Trace	49.9 +/- 1.8	ND	ND	ND	0.2 +/- 0.01	ND
H001979	2015	3	11.9 +/- 0.4	55.3 +/- 3.1	Trace	44.7 +/- 1.6	ND	ND	ND	ND	ND
H001181	2015	4	13.0 +/- 0.4	77.6 +/- 3.9	<0.10	22.3 +/- 1.2	ND	ND	ND	<0.10	ND
H003181	2015	5	13.1 +/- 0.4	83.7 +/- 4.1	10.5 +/- 0.5	5.7 +/- 1.0	ND	<0.10	ND	Trace	Trace
H003258	2015	6	11.2 +/- 0.4	58.1 +/- 3.2	<0.10	41.9 +/- 1.6	ND	Trace	ND	Trace	ND
H003737	2015	7	10.4 +/- 0.4	64.3 +/- 3.4	9.2 +/- 0.4	26.5 +/- 1.3	ND	<0.10	ND	Trace	Trace
H003896	2015	8	21.6 +/- 0.5	32.4 +/- 2.5	47.7 +/- 2	19.8 +/- 1.1	ND	0.1 +/- 0.01	ND	ND	Trace
H004302	2015	9	11.6 +/- 0.4	50.4 +/- 3.0	23.1 +/- 1	26.4 +/- 1.2	ND	0.1 +/- 0.01	ND	ND	Trace
H001191	2016	1	13.0 +/- 0.4	59.3 +/- 3.2	5.5 +/- 0.3	35.2 +/- 1.4	ND	Trace	ND	Trace	ND
H002556	2016	2	10.5 +/- 0.4	63.3 +/- 3.4	0.2 +/- 0.1	36.5 +/- 1.4	ND	<0.10	ND	Trace	ND

3013 Container	FY	DE	Pressure (psia)	He (%)	H ₂ (%)	N ₂ (%)	O ₂ (%)	CO ₂ (%)	CO (%)	N ₂ O (%)	CH ₄ (%)
H004173	2016	3	10.4 +/- 0.4	80.7 +/- 4.0	0.7 +/- 0.1	18.5 +/- 1.2	ND	<0.10	<0.10	Trace	ND
H004247	2016	4	12.0 +/- 0.4	55.7 +/- 3.1	Trace	44.3 +/- 1.6	ND	ND	ND	Trace	ND
H003775	2016	5	10.4 +/- 0.4	60.8 +/- 3.3	<0.10	39.2 +/- 1.5	ND	ND	ND	Trace	Trace
H004024	2016	6	11.1 +/- 0.4	59.6 +/- 3.3	<0.10	40.4 +/- 1.6	ND	Trace	ND	Trace	ND
H001304	2017	1	12.3 +/- 0.4	47.0 +/- 2.9	2.9 +/- 0.2	50.1 +/- 1.8	ND	ND	ND	Trace	ND
H002575	2017	2	13.7 +/- 0.4	45.2 +/- 2.8	22.0 +/- 0.9	32.8 +/- 1.4	ND	Trace	ND	Trace	ND
H003352	2017	3	8.4 +/- 0.4	75.3 +/- 3.8	2.5 +/- 0.2	22.1 +/- 1.2	ND	Trace	ND	Trace	ND
H003695	2017	4	18.7 +/- 0.5	35.2 +/- 2.5	46.6 +/- 2.0	18.1 +/- 1.1	ND	<0.10	ND	ND	<0.10
H002508	2017	5	15.1 +/- 0.4	43.8 +/- 2.8	30.7 +/- 1.3	25.5 +/- 1.2	ND	<0.10	ND	Trace	Trace
R600793	2017	6	13.1 +/- 0.5	90.0 +/- 4.3	Trace	10.0 +/- 1.0	ND	ND	ND	Trace	ND

Appendix 3. The volume of the inner container with convenience container, the material mass and particle density, the material volume, the free gas volume, the wattage of the material, and the best moisture percentage for each container.

3013 Container	FY	DE	Inner container volume (L)	Material mass (g)	Material particle density (g mL⁻³)	Material volume (mL)	Free gas volume (mL)	Material wattage (W)	Average gas temperatur e (K)	Best moisture (%)
R600885	2007	1	1.9780	4313.7	11.560	373.2	1604.8	9.419	341.5	0.1050
R601722	2007	2	1.9780	4410.3	11.000	400.9	1577.1	9.339	341.5	0.1830
R601957	2007	3	1.9780	3877.7	11.340	341.9	1636.1	8.377	341.5	0.0330
R600719	2007	4	1.9780	4352.2	10.620	409.8	1568.2	9.299	341.5	0.1040
R610735	2007	5	1.9780	2373.3	4.500	527.4	1450.6	3.085	341.5	0.3581
R610697	2007	6	1.9780	2832.9	5.950	476.1	1501.9	4.683	341.5	0.2806
R601285	2007	7	1.9780	4733.8	10.950	432.3	1545.7	10.462	341.5	0.1490
R602731	2008	1	1.9780	3443.7	9.420	365.6	1612.4	4.748	341.5	0.0720
R601318	2008	2	1.9780	3960.0	10.410	380.4	1597.6	8.401	341.5	0.1660
H000898	2008	3	1.9090	4137.0	9.150	452.1	1456.9	8.237	341.5	0.0013
R610327	2008	4	1.9780	3647.8	7.240	503.8	1474.2	1.345	341.5	0.0026
R610298	2008	5	1.9780	3801.6	6.030	630.4	1347.6	6.120	341.5	0.0043
R610324	2008	6	1.9780	3042.4	6.690	454.8	1523.2	5.319	341.5	0.0102
H001992	2008	7	1.9090	3185.4	6.890	462.3	1446.7	4.721	341.5	0.0743
H003157	2008	8	1.9090	1709.8	10.960	156.0	1753.0	14.458	341.5	0.1233
R610584	2008	9	1.9780	3490.4	8.540	408.7	1569.3	6.398	341.5	0.1400
R610578	2008	10	1.9780	3532.9	7.220	489.3	1488.7	5.900	341.5	0.0200
H001916	2008	11	1.9090	1896.6	4.960	382.4	1526.6	1.684	341.5	-0.0022
H002088	2008	12	1.9090	1017.4	11.070	91.9	1817.1	11.418	341.5	0.1433
H003409	2008	13	1.9090	2485.2	6.970	356.6	1552.4	4.531	341.5	0.2114
H002573	2008	14	1.9090	2500.2	7.170	348.7	1560.3	4.402	341.5	0.2744

3013 Container	FY	DE	Inner container volume (L)	Material mass (g)	Material particle density (g mL ⁻³)	Material volume (mL)	Free gas volume (mL)	Material wattage (W)	Average gas temperature (K)	Best moisture (%)
H002534	2008	15	1.9090	2500.1	6.960	359.2	1549.8	4.262	341.5	0.2483
R610679	2008	16	1.9780	4536.9	9.060	500.8	1477.2	6.991	341.5	0.0401
H002750	2008	17	1.9090	3135.3	8.060	389.0	1520.0	5.830	341.5	0.0125
H004099	2009	1	1.9090	3435.9	10.090	340.5	1568.5	7.543	341.5	0.2003
H004111	2009	2	1.9090	2483.4	7.130	348.3	1560.7	4.348	341.5	0.2861
H002554	2009	3	1.9090	2506.6	6.760	370.8	1538.2	4.295	341.5	0.2410
H001941	2009	4	1.9090	2801.8	7.800	359.2	1549.8	5.498	341.5	0.0059
R602498	2009	5	1.9780	3378.3	9.920	340.6	1637.4	7.054	341.5	0.2600
H002509	2009	6	1.9090	2487.6	7.040	353.4	1555.6	4.290	341.5	0.3080
H002565	2009	7	1.9090	2493.0	6.490	384.1	1524.9	4.270	341.5	0.2920
H002657	2009	8	1.9090	2500.1	8.540	292.8	1616.2	4.636	341.5	0.1750
R611398	2009	9	1.9780	3337.8	9.070	368.0	1610.0	5.947	341.5	0.2887
H002200	2009	10	1.9090	4037.1	8.330	484.6	1424.4	7.709	341.5	0.0210
H002667	2009	11	1.9090	2331.6	6.760	344.9	1564.1	4.082	341.5	0.1555
H002715	2009	12	1.9090	2493.5	6.420	388.4	1520.6	4.270	341.5	0.3090
R610700	2009	13	1.9780	4540.4	8.380	541.8	1436.2	7.534	341.5	0.0253
R610764	2009	14	1.9780	1996.2	6.210	321.4	1656.6	3.055	341.5	0.0477
R610573	2009	15	1.9780	2799.2	6.960	402.2	1575.8	4.459	341.5	0.1213
R610558	2009	16	1.9780	3950.9	11.110	355.6	1622.4	8.510	341.5	-0.0017
R610806	2009	17	1.9780	4366.5	8.650	504.8	1473.2	8.193	341.5	0.1116
H003119	2009	18	1.9090	2429.0	7.470	325.2	1583.8	5.532	341.5	0.0967
H002195	2009	19	1.9090	3937.1	9.630	408.8	1500.2	8.770	341.5	0.0000
H004251	2010	1	1.9090	2578.4	4.920	524.1	1384.9	3.745	341.5	0.1777
H002496	2010	2	1.9090	1886.5	6.060	311.3	1597.7	2.470	341.5	0.2072
H003710	2010	3	1.9090	2498.6	6.840	365.3	1543.7	4.460	341.5	0.3360

3013 Container	FY	DE	Inner container volume (L)	Material mass (g)	Material particle density (g mL ⁻³)	Material volume (mL)	Free gas volume (mL)	Material wattage (W)	Average gas temperature (K)	Best moisture (%)
H003655	2010	4	1.9090	2588.8	6.610	391.6	1517.4	4.541	341.5	0.2575
H002447	2010	5	1.9090	2435.1	9.250	263.3	1645.7	4.765	341.5	0.0384
R610627	2010	6	1.9780	3286.3	4.700	699.2	1278.8	4.401	341.5	0.0163
H003900	2010	7	1.9090	2479.9	5.430	456.7	1452.3	3.658	341.5	0.3037
H003650	2010	8	1.9090	2483.2	7.770	319.6	1589.4	4.623	341.5	0.2921
H002567	2010	9	1.9090	814.6	6.250	130.3	1778.7	1.232	341.5	0.0436
H002728	2010	10	1.9090	2474.5	6.640	372.7	1536.3	4.347	341.5	0.1631
H002786	2010	11	1.9090	2504.8	6.650	376.7	1532.3	4.290	341.5	0.2468
H003077	2010	12	1.9090	3332.5	10.180	327.4	1581.6	6.860	341.5	0.0873
H003367	2010	13	1.9090	2052.0	6.040	339.7	1569.3	2.675	341.5	0.1319
H003704	2010	14	1.9090	2488.7	6.340	392.5	1516.5	4.443	341.5	0.2820
R610785	2010	15	1.9780	3023.5	5.800	521.3	1456.7	4.658	341.5	0.0117
R610826	2010	16	1.9780	3532.0	4.700	751.5	1226.5	4.694	341.5	0.0079
R610853	2010	17	1.9780	4116.6	7.380	557.8	1420.2	7.167	341.5	0.0269
S001721	2010	18	1.9940	3971.3	10.420	381.1	1612.9	8.319	341.5	0.1900
H003443	2011	1	1.9090	2491.6	6.960	358.0	1551.0	4.224	341.5	0.2968
S002129	2011	2	1.9940	4737.1	9.950	476.1	1517.9	9.633	341.5	0.2400
H002592	2011	3	1.9090	1374.9	6.400	214.8	1694.2	1.818	341.5	0.0304
H003337	2011	4	1.9090	2392.4	5.160	463.6	1445.4	3.272	341.5	0.1123
S001105	2011	5	1.9940	4814.4	10.150	474.3	1519.7	9.721	341.5	0.1967
H003343	2011	6	1.9090	1921.9	8.210	234.1	1674.9	3.630	341.5	0.1712
H003371	2011	7	1.9090	2233.9	6.970	320.5	1588.5	3.027	341.5	0.1784
H003526	2011	8	1.9090	2491.9	6.860	363.3	1545.7	4.272	341.5	0.1319
H003565	2011	9	1.9090	2466.9	7.920	311.5	1597.5	4.619	341.5	0.1588
R611131	2011	10	1.9780	3603.9	8.180	440.6	1537.4	6.984	341.5	0.0147

3013 Container	FY	DE	Inner container volume (L)	Material mass (g)	Material particle density (g mL ⁻³)	Material volume (mL)	Free gas volume (mL)	Material wattage (W)	Average gas temperature (K)	Best moisture (%)
H003625	2011	11	1.9090	2484.4	7.440	333.9	1575.1	4.580	341.5	0.1399
L000178	2011	12	1.9780	2714.9	6.090	445.8	1532.2	2.961	341.5	0.0060
H003328	2011	13	1.9090	2484.9	8.060	308.3	1600.7	4.920	341.5	0.5153
H001209	2012	1	1.9090	790.2	6.020	131.3	1777.7	1.120	341.5	0.2772
H002574	2012	2	1.9090	1909.7	5.430	351.7	1557.3	2.383	341.5	0.1403
H001513	2012	3	1.9090	827.2	6.170	134.1	1774.9	1.192	341.5	0.3073
H003390	2012	4	1.9090	3135.7	7.800	402.0	1507.0	4.707	341.5	0.1137
L000075	2012	5	1.9780	4847.8	10.550	459.5	1518.5	9.778	341.5	0.0005
H004012	2012	6	1.9090	2490.9	5.620	443.2	1465.8	3.840	341.5	0.1447
H004048	2012	7	1.9090	2433.8	4.730	514.5	1394.5	3.338	341.5	0.1017
R610960	2012	8	1.9780	3211.3	5.150	623.6	1354.4	4.596	341.5	0.0343
S002250	2012	10	1.9940	4474.0	8.580	521.4	1472.6	6.693	341.5	0.0233
H001236	2013	1	1.9090	739.1	4.860	152.1	1756.9	0.840	299.0	0.4000
R610996	2014	1	1.9780	3787.8	4.750	797.4	1180.6	5.440	341.5	0.0105
H003064	2014	2	1.9090	2487.1	6.170	403.1	1505.9	4.145	341.5	0.3001
H003307	2014	3	1.9090	2277.0	5.900	385.9	1523.1	3.739	313.4	0.1694
H003052	2014	4	1.9090	2504.7	6.319	396.4	1512.6	4.180	312.7	0.1942
H003898	2014	5	1.9090	2480.0	5.656	438.5	1470.5	3.972	311.4	0.1594
S002277	2014	6	1.9940	4781.6	10.114	472.8	1521.2	9.587	333.0	0.2233
S002116	2014	7	1.9940	4778.4	9.979	478.8	1515.2	9.488	333.8	0.1333
H004219	2014	8	1.9090	2080.5	6.011	346.1	1562.9	3.405	309.7	0.1584
H002636	2014	9	1.9090	2888.0	10.055	287.2	1621.8	9.458	333.8	0.0085
R610156	2015	1	1.9780	1234.3	4.434	278.4	1699.6	1.541	300.9	0.3200
S002162	2015	2	1.9940	4742.3	10.383	456.8	1537.2	9.559	332.7	0.0967
H001979	2015	3	1.9090	2362.2	6.958	339.5	1569.5	4.208	316.0	0.0977

3013 Container	FY	DE	Inner container volume (L)	Material mass (g)	Material particle density (g mL ⁻³)	Material volume (mL)	Free gas volume (mL)	Material wattage (W)	Average gas temperatur e (K)	Best moisture (%)
H001181	2015	4	1.9090	899.3	7.647	117.6	1791.4	1.740	300.6	0.3300
H003181	2015	5	1.9090	2492.3	7.943	313.8	1595.2	4.760	314.3	0.2849
H003258	2015	6	1.9090	2483.6	7.192	345.3	1563.7	4.304	312.3	0.1469
H003737	2015	7	1.9090	2483.5	6.409	387.5	1521.5	4.280	313.6	0.2400
H003896	2015	8	1.9090	2541.5	5.876	432.5	1476.5	3.910	310.7	0.2826
H004302	2015	9	1.9090	2425.8	5.947	407.9	1501.1	4.230	312.4	0.1926
H001191	2016	1	1.9090	993.4	7.699	129.0	1780.0	1.840	303.8	0.3056
H002556	2016	2	1.9090	2518.2	7.663	328.6	1580.4	4.497	314.3	0.1500
H004173	2016	3	1.9090	2492.3	6.213	401.1	1507.9	4.390	313.2	0.1690
H004247	2016	4	1.9090	2483.0	5.976	415.5	1493.5	4.040	311.4	0.1305
H003775	2016	5	1.9090	2466.7	7.723	319.4	1589.6	5.320	316.9	0.1487
H004024	2016	6	1.9090	2476.6	6.599	375.3	1533.7	4.360	313.0	0.1865
H001304	2017	1	1.9090	1034.9	4.956	208.8	1700.2	1.174	299.5	0.2670
H002575	2017	2	1.9090	2488.8	7.830	317.8	1591.2	4.580	313.9	0.2960
H003352	2017	3	1.9090	2438.8	7.326	332.9	1576.1	4.544	314.3	0.2900
H003695	2017	4	1.9090	2499.9	6.769	369.3	1539.7	4.450	313.9	0.2650
H002508	2017	5	1.9090	2459.8	6.213	395.9	1513.1	4.180	311.8	0.2420
R600793	2017	6	1.9780	4522.1	9.686	466.9	1511.1	9.722	336.4	0.3200

Appendix 4. The amount of each gas calculated from the pressure, the gas volume, and the measured percent of each gas (input data from Appendix 2 and Appendix 3) Reported percentages that were non-numeric (ND, Trace, and <0.01) are reported as --- in this table.

3013 Container	FY	DE	He (mole)	H₂ (mole)	N₂ (mole)	O₂ (mole)	CO₂ (mole)	CO (mole)	N₂O (mole)	CH₄ (mole)
R600885	2007	1	4.53E-02	---	4.59E-03	---	---	---	---	---
R601722	2007	2	4.17E-02	---	6.08E-03	---	---	---	---	---
R601957	2007	3	4.56E-02	---	5.29E-03	---	---	---	---	---
R600719	2007	4	4.12E-02	---	7.96E-03	---	---	---	---	---
R610735	2007	5	3.60E-02	---	7.64E-03	---	9.61E-05	---	---	---
R610697	2007	6	3.74E-02	---	8.21E-03	---	---	---	---	---
R601285	2007	7	3.89E-02	---	1.13E-02	---	---	---	---	---
R602731	2008	1	4.04E-02	---	7.75E-03	---	---	---	---	---
R601318	2008	2	4.47E-02	---	4.16E-03	---	---	---	---	---
H000898	2008	3	3.49E-02	---	8.40E-03	---	1.31E-04	---	---	---
R610327	2008	4	3.68E-02	---	7.41E-03	---	1.78E-04	---	---	---
R610298	2008	5	3.57E-02	---	6.81E-03	---	---	---	---	---
R610324	2008	6	3.99E-02	---	8.58E-03	---	---	---	---	---
H001992	2008	7	2.70E-02	---	1.97E-02	---	---	---	---	---
H003157	2008	8	2.87E-02	---	2.32E-02	8.97E-04	---	---	---	---
R610584	2008	9	4.42E-02	---	7.73E-03	---	3.65E-04	---	---	---
R610578	2008	10	3.71E-02	---	8.04E-03	---	---	---	---	---
H001916	2008	11	2.69E-02	---	2.25E-02	---	---	---	---	---
H002088	2008	12	3.20E-02	---	2.28E-02	---	---	---	---	---
H003409	2008	13	2.38E-02	9.45E-03	1.73E-02	---	---	---	---	---
H002573	2008	14	2.53E-02	1.86E-02	2.26E-02	---	2.00E-04	---	---	---
H002534	2008	15	2.13E-02	1.59E-02	1.24E-02	---	1.39E-03	4.12E-04	---	2.06E-04
R610679	2008	16	2.95E-02	---	1.12E-02	---	1.26E-03	---	---	---

3013 Container	FY	DE	He (mole)	H ₂ (mole)	N ₂ (mole)	O ₂ (mole)	CO ₂ (mole)	CO (mole)	N ₂ O (mole)	CH ₄ (mole)
H002750	2008	17	3.05E-02	---	1.78E-02	4.84E-05	---	---	---	---
H004099	2009	1	2.01E-02	---	2.36E-02	8.91E-05	1.78E-04	---	6.24E-04	---
H004111	2009	2	1.86E-02	1.17E-02	2.42E-02	---	---	---	---	---
H002554	2009	3	2.10E-02	1.02E-02	1.51E-02	---	---	---	---	---
H001941	2009	4	2.49E-02	---	2.47E-02	---	---	---	---	---
R602498	2009	5	4.28E-02	---	6.11E-03	---	---	---	---	---
H002509	2009	6	2.65E-02	2.83E-02	2.27E-02	---	3.11E-04	---	---	---
H002565	2009	7	2.29E-02	1.81E-03	1.64E-02	---	---	---	---	---
H002657	2009	8	2.72E-02	3.08E-04	1.65E-02	---	---	---	---	---
R611398	2009	9	3.59E-02	---	8.42E-03	---	---	---	1.78E-04	---
H002200	2009	10	2.36E-02	---	2.31E-02	---	---	---	---	---
H002667	2009	11	2.37E-02	---	2.04E-02	---	---	---	---	---
H002715	2009	12	2.60E-02	2.91E-02	1.83E-02	---	1.97E-03	1.51E-04	---	7.57E-05
R610700	2009	13	3.33E-02	---	5.70E-03	---	1.16E-03	---	---	---
R610764	2009	14	4.09E-02	---	8.25E-03	---	---	---	---	---
R610573	2009	15	4.48E-02	---	7.44E-03	---	---	---	1.57E-04	---
R610558	2009	16	3.96E-02	---	8.34E-03	4.85E-05	1.45E-04	---	2.91E-04	---
R610806	2009	17	3.68E-02	---	6.59E-03	---	---	---	2.62E-04	---
H003119	2009	18	2.55E-02	---	1.95E-02	---	---	---	9.00E-05	---
H002195	2009	19	2.28E-02	---	2.72E-02	---	---	---	---	---
H004251	2010	1	3.14E-02	1.61E-02	9.54E-03	---	1.47E-03	2.35E-04	---	1.18E-04
H002496	2010	2	2.44E-02	5.85E-04	2.00E-02	---	---	---	---	---
H003710	2010	3	2.48E-02	1.60E-02	2.12E-02	---	2.49E-04	---	---	---
H003655	2010	4	2.79E-02	1.78E-02	1.90E-02	---	1.95E-04	---	---	---
H002447	2010	5	2.49E-02	---	2.28E-02	4.80E-05	---	---	1.44E-04	---
R610627	2010	6	3.88E-02	---	6.76E-03	---	---	---	---	---
H003900	2010	7	2.64E-02	1.80E-02	1.53E-02	---	1.20E-04	---	---	---

3013 Container	FY	DE	He (mole)	H ₂ (mole)	N ₂ (mole)	O ₂ (mole)	CO ₂ (mole)	CO (mole)	N ₂ O (mole)	CH ₄ (mole)
H003650	2010	8	2.58E-02	8.35E-03	1.53E-02	---	---	---	---	---
H002567	2010	9	2.93E-02	---	2.13E-02	---	---	---	---	---
H002728	2010	10	2.30E-02	6.26E-03	1.33E-02	-2.55E-05	---	---	---	---
H002786	2010	11	2.41E-02	4.90E-03	1.75E-02	---	---	---	---	---
H003077	2010	12	2.45E-02	---	2.19E-02	---	---	---	4.55E-04	---
H003367	2010	13	2.68E-02	3.42E-04	1.66E-02	---	---	---	---	---
H003704	2010	14	2.33E-02	1.27E-02	2.03E-02	---	---	---	---	---
R610785	2010	15	4.17E-02	3.44E-04	7.13E-03	---	---	---	---	---
R610826	2010	16	3.16E-02	---	4.11E-03	---	---	---	---	---
R610853	2010	17	3.68E-02	---	5.60E-03	---	---	---	---	---
S001721	2010	18	3.46E-02	---	1.23E-02	---	---	---	4.70E-05	---
H003443	2011	1	2.43E-02	2.50E-02	1.88E-02	---	---	---	---	---
S002129	2011	2	2.74E-02	---	1.81E-02	---	---	---	1.83E-04	---
H002592	2011	3	2.91E-02	---	1.94E-02	---	---	---	---	---
H003337	2011	4	2.71E-02	---	1.61E-02	---	---	---	---	---
S001105	2011	5	2.00E-02	---	2.39E-02	---	---	---	3.99E-04	---
H003343	2011	6	2.61E-02	---	2.09E-02	---	---	---	2.36E-04	---
H003371	2011	7	2.65E-02	7.05E-03	1.32E-02	---	---	---	---	---
H003526	2011	8	2.46E-02	1.88E-04	1.28E-02	---	---	---	---	---
H003565	2011	9	2.77E-02	3.67E-04	1.27E-02	---	---	---	---	---
R611131	2011	10	4.44E-02	---	7.16E-03	---	---	---	---	---
H003625	2011	11	3.53E-02	2.26E-04	9.61E-03	---	---	---	---	---
L000178	2011	12	1.18E-03	---	5.02E-02	---	---	---	---	---
H003328	2011	13	2.39E-02	1.25E-01	1.91E-02	1.18E-04	---	---	---	---
H001209	2012	1	2.87E-02	2.09E-04	2.33E-02	---	---	---	---	---
H002574	2012	2	4.44E-02	---	3.92E-03	---	---	---	---	---
H001513	2012	3	2.57E-02	---	2.26E-02	---	---	---	2.44E-04	---

3013 Container	FY	DE	He (mole)	H ₂ (mole)	N ₂ (mole)	O ₂ (mole)	CO ₂ (mole)	CO (mole)	N ₂ O (mole)	CH ₄ (mole)
H003390	2012	4	2.41E-02	8.34E-05	1.75E-02	---	---	---	8.34E-05	---
L000075	2012	5	9.64E-03	---	4.04E-02	---	1.83E-03	---	4.71E-04	---
H004012	2012	6	1.88E-02	---	2.00E-02	---	---	---	---	---
H004048	2012	7	1.72E-02	1.36E-03	1.44E-02	---	1.33E-04	---	---	---
R610960	2012	8	3.74E-02	5.00E-03	3.42E-04	---	---	---	---	---
S002250	2012	10	3.56E-02	---	1.05E-02	4.86E-04	1.75E-03	1.46E-04	1.95E-04	---
H001236	2013	1	3.05E-02	4.23E-04	2.95E-02	---	---	---	---	---
R610996	2014	1	3.24E-02	---	2.59E-03	---	---	---	---	---
H003064	2014	2	3.82E-02	3.68E-02	8.54E-03	---	1.67E-04	---	---	---
H003307	2014	3	2.46E-02	---	2.05E-02	---	---	---	---	---
H003052	2014	4	2.72E-02	3.30E-03	1.12E-02	---	---	---	---	---
H003898	2014	5	2.46E-02	7.65E-03	1.35E-02	---	4.58E-05	---	---	---
S002277	2014	6	2.14E-02	---	2.47E-02	---	---	---	9.24E-05	---
S002116	2014	7	3.92E-02	---	9.69E-03	---	---	---	---	---
H004219	2014	8	3.03E-02	3.96E-04	1.33E-02	---	---	---	---	---
H002636	2014	9	2.52E-02	---	2.43E-02	---	---	---	---	---
R610156	2015	1	4.87E-02	---	9.84E-03	---	---	---	---	---
S002162	2015	2	2.31E-02	---	2.31E-02	---	---	---	9.27E-05	---
H001979	2015	3	2.71E-02	---	2.19E-02	---	---	---	---	---
H001181	2015	4	4.99E-02	---	1.43E-02	---	---	---	---	---
H003181	2015	5	4.62E-02	5.79E-03	3.14E-03	---	---	---	---	---
H003258	2015	6	2.70E-02	---	1.95E-02	---	---	---	---	---
H003737	2015	7	2.69E-02	3.85E-03	1.11E-02	---	---	---	---	---
H003896	2015	8	2.76E-02	4.06E-02	1.69E-02	---	8.51E-05	---	---	---
H004302	2015	9	2.33E-02	1.07E-02	1.22E-02	---	4.62E-05	---	---	---
H001191	2016	1	3.75E-02	3.47E-03	2.22E-02	---	---	---	---	---
H002556	2016	2	2.77E-02	8.76E-05	1.60E-02	---	---	---	---	---

3013 Container	FY	DE	He (mole)	H₂ (mole)	N₂ (mole)	O₂ (mole)	CO₂ (mole)	CO (mole)	N₂O (mole)	CH₄ (mole)
H004173	2016	3	3.35E-02	2.91E-04	7.68E-03	---	---	---	---	---
H004247	2016	4	2.66E-02	---	2.11E-02	---	---	---	---	---
H003775	2016	5	2.63E-02	---	1.70E-02	---	---	---	---	---
H004024	2016	6	2.69E-02	---	1.82E-02	---	---	---	---	---
H001304	2017	1	2.72E-02	1.68E-03	2.90E-02	---	---	---	---	---
H002575	2017	2	2.60E-02	1.27E-02	1.89E-02	---	---	---	---	---
H003352	2017	3	2.63E-02	8.89E-04	7.72E-03	---	---	---	---	---
H003695	2017	4	2.68E-02	3.54E-02	1.38E-02	---	---	---	---	---
H002508	2017	5	2.66E-02	1.87E-02	1.55E-02	---	---	---	---	---
R600793	2017	6	4.39E-02	---	4.88E-03	---	---	---	---	---

Appendix 5. The calculated gas composition and the total pressure at the time the inner can was welded. In order to calculate the initial composition, 25% of the He generated during storage is subtracted from the amount of He observed at DE. The generated He typically represents a small fraction of the amount of He observed at DE.

3013 Container	FY	DE	He generated (mole)	Fraction of final He	He (mole)	N₂ (mole)	O₂ (mole)	total (mole)	Pressure (kPa)
R600885	2007	1	0.00311	0.069	0.0445	0.0046	0.0012	0.0503	89.1
R601722	2007	2	0.00312	0.075	0.0410	0.0061	0.0016	0.0487	87.6
R601957	2007	3	0.00282	0.062	0.0449	0.0053	0.0014	0.0516	89.5
R600719	2007	4	0.00320	0.078	0.0404	0.0080	0.0021	0.0505	91.4
R610735	2007	5	0.00081	0.023	0.0357	0.0076	0.0021	0.0454	88.9
R610697	2007	6	0.00124	0.033	0.0371	0.0082	0.0022	0.0475	89.8
R601285	2007	7	0.00345	0.089	0.0381	0.0113	0.0030	0.0524	96.3
R602731	2008	1	0.00151	0.037	0.0400	0.0078	0.0021	0.0499	87.8
R601318	2008	2	0.00289	0.065	0.0440	0.0042	0.0011	0.0493	87.6
H000898	2008	3	0.00256	0.073	0.0343	0.0084	0.0023	0.0450	87.6
R610327	2008	4	0.00042	0.011	0.0367	0.0074	0.0020	0.0461	88.7
R610298	2008	5	0.00192	0.054	0.0353	0.0068	0.0018	0.0439	92.5
R610324	2008	6	0.00164	0.041	0.0395	0.0086	0.0023	0.0503	93.8
H001992	2008	7	0.00161	0.059	0.0266	0.0197	0.0000	0.0463	90.9
H003157	2008	8	0.00486	0.169	0.0275	0.0232	0.0062	0.0569	92.2
R610584	2008	9	0.00199	0.045	0.0437	0.0077	0.0021	0.0535	96.7
R610578	2008	10	0.00185	0.050	0.0366	0.0080	0.0022	0.0468	89.3
H001916	2008	11	0.00060	0.022	0.0267	0.0225	0.0000	0.0492	91.5
H002088	2008	12	0.00394	0.123	0.0310	0.0228	0.0061	0.0598	93.5
H003409	2008	13	0.00140	0.059	0.0234	0.0173	0.0046	0.0454	83.0
H002573	2008	14	0.00137	0.054	0.0249	0.0226	0.0061	0.0536	97.5

3013 Container	FY	DE	He generated (mole)	Fraction of final He	He (mole)	N ₂ (mole)	O ₂ (mole)	total (mole)	Pressure (kPa)
H002534	2008	15	0.00133	0.063	0.0210	0.0124	0.0033	0.0367	67.2
R610679	2008	16	0.00231	0.078	0.0289	0.0112	0.0030	0.0431	82.9
H002750	2008	17	0.00186	0.061	0.0300	0.0178	0.0000	0.0478	89.4
H004099	2009	1	0.00225	0.112	0.0195	0.0242	0.0065	0.0502	90.9
H004111	2009	2	0.00131	0.070	0.0183	0.0242	0.0065	0.0490	89.2
H002554	2009	3	0.00141	0.067	0.0206	0.0151	0.0041	0.0398	73.4
H001941	2009	4	0.00210	0.084	0.0244	0.0247	0.0000	0.0492	90.0
R602498	2009	5	0.00315	0.074	0.0420	0.0061	0.0016	0.0498	86.3
H002509	2009	6	0.00147	0.055	0.0262	0.0227	0.0061	0.0550	100.4
H002565	2009	7	0.00147	0.064	0.0226	0.0164	0.0044	0.0433	80.7
H002657	2009	8	0.00162	0.060	0.0268	0.0165	0.0044	0.0477	83.7
R611398	2009	9	0.00211	0.059	0.0354	0.0086	0.0023	0.0463	81.7
H002200	2009	10	0.00295	0.125	0.0228	0.0231	0.0000	0.0460	91.6
H002667	2009	11	0.00146	0.062	0.0233	0.0204	0.0055	0.0491	89.2
H002715	2009	12	0.00152	0.058	0.0257	0.0183	0.0049	0.0489	91.3
R610700	2009	13	0.00284	0.085	0.0325	0.0057	0.0015	0.0398	78.6
R610764	2009	14	0.00116	0.028	0.0406	0.0082	0.0022	0.0511	87.5
R610573	2009	15	0.00171	0.038	0.0444	0.0076	0.0020	0.0540	97.4
R610558	2009	16	0.00331	0.083	0.0388	0.0086	0.0023	0.0498	87.1
R610806	2009	17	0.00320	0.087	0.0360	0.0069	0.0018	0.0447	86.1
H003119	2009	18	0.00191	0.075	0.0250	0.0196	0.0053	0.0498	89.3
H002195	2009	19	0.00357	0.157	0.0219	0.0272	0.0000	0.0490	92.8
H004251	2010	1	0.00135	0.043	0.0311	0.0095	0.0026	0.0432	88.5
H002496	2010	2	0.00095	0.039	0.0242	0.0200	0.0054	0.0495	88.0
H003710	2010	3	0.00171	0.069	0.0243	0.0212	0.0057	0.0512	94.1
H003655	2010	4	0.00176	0.063	0.0274	0.0190	0.0051	0.0515	96.5

3013 Container	FY	DE	He generated (mole)	Fraction of final He	He (mole)	N ₂ (mole)	O ₂ (mole)	total (mole)	Pressure (kPa)
H002447	2010	5	0.00195	0.078	0.0245	0.0230	0.0062	0.0536	92.4
R610627	2010	6	0.00182	0.047	0.0384	0.0068	0.0018	0.0470	104.3
H003900	2010	7	0.00142	0.054	0.0261	0.0153	0.0041	0.0455	88.9
H003650	2010	8	0.00185	0.072	0.0253	0.0153	0.0041	0.0447	79.8
H002567	2010	9	0.00050	0.017	0.0291	0.0213	0.0057	0.0561	89.5
H002728	2010	10	0.00182	0.079	0.0225	0.0133	0.0036	0.0394	72.8
H002786	2010	11	0.00180	0.075	0.0236	0.0175	0.0047	0.0458	84.9
H003077	2010	12	0.00294	0.120	0.0238	0.0224	0.0060	0.0521	93.5
H003367	2010	13	0.00115	0.043	0.0265	0.0166	0.0045	0.0476	86.2
H003704	2010	14	0.00186	0.080	0.0228	0.0203	0.0054	0.0485	90.9
R610785	2010	15	0.00207	0.050	0.0412	0.0071	0.0019	0.0502	97.9
R610826	2010	16	0.00209	0.066	0.0311	0.0041	0.0011	0.0363	84.1
R610853	2010	17	0.00321	0.087	0.0360	0.0056	0.0015	0.0431	86.2
S001721	2010	18	0.00321	0.093	0.0338	0.0124	0.0033	0.0495	87.1
H003443	2011	1	0.00187	0.077	0.0239	0.0188	0.0050	0.0477	87.4
S002129	2011	2	0.00355	0.130	0.0265	0.0183	0.0049	0.0497	93.0
H002592	2011	3	0.00081	0.028	0.0289	0.0194	0.0052	0.0535	89.6
H003337	2011	4	0.00143	0.053	0.0267	0.0161	0.0043	0.0471	92.5
S001105	2011	5	0.00366	0.183	0.0191	0.0243	0.0065	0.0499	93.2
H003343	2011	6	0.00162	0.062	0.0257	0.0211	0.0057	0.0524	88.9
H003371	2011	7	0.00142	0.054	0.0261	0.0132	0.0035	0.0428	76.5
H003526	2011	8	0.00197	0.080	0.0241	0.0128	0.0034	0.0403	74.0
H003565	2011	9	0.00213	0.077	0.0271	0.0127	0.0034	0.0432	76.8
R611131	2011	10	0.00337	0.076	0.0435	0.0072	0.0019	0.0526	97.1
H003625	2011	11	0.00215	0.061	0.0348	0.0096	0.0026	0.0470	84.6
L000178	2011	12	0.00146	1.233	0.0008	0.0502	0.0000	0.0510	94.5

3013 Container	FY	DE	He generated (mole)	Fraction of final He	He (mole)	N ₂ (mole)	O ₂ (mole)	total (mole)	Pressure (kPa)
H003328	2011	13	0.00213	0.089	0.0234	0.0191	0.0000	0.0425	75.4
H001209	2012	1	0.00065	0.023	0.0286	0.0233	0.0063	0.0581	92.8
H002574	2012	2	0.00123	0.028	0.0441	0.0039	0.0011	0.0491	89.5
H001513	2012	3	0.00069	0.027	0.0255	0.0229	0.0061	0.0546	87.3
H003390	2012	4	0.00249	0.103	0.0235	0.0176	0.0047	0.0457	86.1
L000075	2012	5	0.00547	3.603	0.0002 He 0.00812 Ar	0.0409	0.0000	0.0492	91.9
H004012	2012	6	0.00197	0.105	0.0183	0.0200	0.0054	0.0436	84.5
H004048	2012	7	0.00173	0.101	0.0168	0.0144	0.0039	0.0351	71.5
R610960	2012	8	0.00257	0.069	0.0367	0.0003	0.0001	0.0372	77.9
S002250	2012	10	0.00299	0.084	0.0349	0.0107	0.0029	0.0484	93.2
H001236	2013	1	0.00058	0.019	0.0304	0.0295	0.0079	0.0678	95.9
R610996	2014	1	0.00362	0.112	0.0314	0.0026	0.0007	0.0347	83.5
H003064	2014	2	0.00260	0.068	0.0375	0.0085	0.0023	0.0484	91.2
H003307	2014	3	0.00238	0.096	0.0241	0.0205	0.0055	0.0500	85.6
H003052	2014	4	0.00267	0.098	0.0266	0.0112	0.0030	0.0408	70.1
H003898	2014	5	0.00259	0.105	0.0240	0.0135	0.0036	0.0411	72.4
S002277	2014	6	0.00567	0.264	0.0200	0.0248	0.0066	0.0514	93.6
S002116	2014	7	0.00561	0.143	0.0378	0.0097	0.0026	0.0501	91.8
H004219	2014	8	0.00221	0.073	0.0297	0.0133	0.0036	0.0466	76.7
H002636	2014	9	0.00644	0.255	0.0236	0.0243	0.0000	0.0479	82.0
R610156	2015	1	0.00113	0.023	0.0484	0.0098	0.0026	0.0609	89.7
S002162	2015	2	0.00600	0.259	0.0216	0.0232	0.0062	0.0511	91.9
H001979	2015	3	0.00289	0.107	0.0264	0.0219	0.0059	0.0542	90.7
H001181	2015	4	0.00139	0.028	0.0495	0.0143	0.0038	0.0677	94.4
H003181	2015	5	0.00329	0.071	0.0453	0.0031	0.0008	0.0493	80.8

3013 Container	FY	DE	He generated (mole)	Fraction of final He	He (mole)	N ₂ (mole)	O ₂ (mole)	total (mole)	Pressure (kPa)
H003258	2015	6	0.00302	0.112	0.0263	0.0195	0.0052	0.0510	84.6
H003737	2015	7	0.00312	0.116	0.0261	0.0111	0.0030	0.0402	68.9
H003896	2015	8	0.00283	0.102	0.0269	0.0169	0.0045	0.0483	84.4
H004302	2015	9	0.00301	0.129	0.0225	0.0122	0.0033	0.0380	65.8
H001191	2016	1	0.00156	0.042	0.0371	0.0222	0.0060	0.0653	92.6
H002556	2016	2	0.00355	0.128	0.0268	0.0160	0.0043	0.0471	77.9
H004173	2016	3	0.00340	0.102	0.0327	0.0077	0.0021	0.0424	73.2
H004247	2016	4	0.00315	0.118	0.0258	0.0211	0.0057	0.0526	91.2
H003775	2016	5	0.00421	0.160	0.0253	0.0170	0.0045	0.0468	77.5
H004024	2016	6	0.00344	0.128	0.0260	0.0182	0.0049	0.0491	83.4
H001304	2017	1	0.00107	0.039	0.0269	0.0290	0.0078	0.0637	93.4
H002575	2017	2	0.00385	0.148	0.0251	0.0189	0.0051	0.0490	80.4
H003352	2017	3	0.00382	0.145	0.0254	0.0077	0.0021	0.0352	58.3
H003695	2017	4	0.00378	0.141	0.0258	0.0138	0.0037	0.0433	73.4
H002508	2017	5	0.00360	0.135	0.0257	0.0155	0.0042	0.0454	77.7
R600793	2017	6	0.00924	0.210	0.0416	0.0049	0.0013	0.0478	88.5

Appendix 6. The amount of H₂ observed, the amount of H₂ calculated using the 3013 pressure equation, and the hydrogen fraction (ratio of observed to calculated).

3013 Container	FY	DE	H₂ observed (mole)	H₂ calculated using pressure equation (mole)	H₂ Fraction
R600885	2007	1	4.99E-06	0.251	1.98E-05
R601722	2007	2	4.79E-06	0.448	1.07E-05
R601957	2007	3	5.09E-06	0.071	7.16E-05
R600719	2007	4	4.91E-06	0.251	1.96E-05
R610735	2007	5	4.37E-06	0.472	9.26E-06
R610697	2007	6	4.56E-06	0.441	1.03E-05
R601285	2007	7	5.03E-06	0.392	1.28E-05
R602731	2008	1	4.82E-06	0.138	3.50E-05
R601318	2008	2	4.89E-06	0.365	1.34E-05
H000898	2008	3	4.35E-06	0.003	1.45E-03
R610327	2008	4	4.44E-06	0.005	8.46E-04
R610298	2008	5	4.25E-06	0.009	4.70E-04
R610324	2008	6	4.85E-06	0.017	2.80E-04
H001992	2008	7	4.67E-06	0.131	3.56E-05
H003157	2008	8	5.28E-06	0.117	4.51E-05
R610584	2008	9	5.22E-06	0.271	1.92E-05
R610578	2008	10	4.52E-06	0.039	1.15E-04
H001916	2008	11	4.93E-06	0.001	4.68E-03
H002088	2008	12	5.47E-06	0.081	6.76E-05
H003409	2008	13	9.45E-03	0.292	3.24E-02
H002573	2008	14	1.86E-02	0.381	4.89E-02
H002534	2008	15	1.59E-02	0.345	4.61E-02
R610679	2008	16	4.20E-06	0.101	4.16E-05
H002750	2008	17	4.84E-06	0.022	2.23E-04
H004099	2009	1	4.46E-06	0.382	1.17E-05
H004111	2009	2	1.17E-02	0.394	2.98E-02
H002554	2009	3	1.02E-02	0.335	3.05E-02
H001941	2009	4	4.97E-06	0.009	5.38E-04
R602498	2009	5	4.89E-07	0.488	1.00E-06
H002509	2009	6	2.83E-02	0.425	6.64E-02
H002565	2009	7	1.81E-03	0.404	4.48E-03
H002657	2009	8	3.08E-04	0.243	1.27E-03
R611398	2009	9	4.46E-05	0.535	8.33E-05

3013 Container	FY	DE	H₂ observed (mole)	H₂ calculated using pressure equation (mole)	H₂ Fraction
H002200	2009	10	4.67E-05	0.047	9.91E-04
H002667	2009	11	4.41E-05	0.201	2.19E-04
H002715	2009	12	2.91E-02	0.428	6.82E-02
R610700	2009	13	4.01E-05	0.064	6.30E-04
R610764	2009	14	4.91E-05	0.053	9.29E-04
R610573	2009	15	5.24E-06	0.188	2.78E-05
R610558	2009	16	4.85E-06	0.002	2.21E-03
R610806	2009	17	4.36E-05	0.271	1.61E-04
H003119	2009	18	4.50E-05	0.130	3.45E-04
H002195	2009	19	4.99E-06	0.002	2.28E-03
H004251	2010	1	1.61E-02	0.254	6.34E-02
H002496	2010	2	5.85E-04	0.217	2.70E-03
H003710	2010	3	1.60E-02	0.466	3.43E-02
H003655	2010	4	1.78E-02	0.370	4.80E-02
H002447	2010	5	4.80E-05	0.052	9.24E-04
R610627	2010	6	4.57E-06	0.030	1.53E-04
H003900	2010	7	1.80E-02	0.418	4.32E-02
H003650	2010	8	8.35E-03	0.403	2.07E-02
H002567	2010	9	5.05E-05	0.020	2.56E-03
H002728	2010	10	6.26E-03	0.224	2.79E-02
H002786	2010	11	4.90E-03	0.343	1.43E-02
H003077	2010	12	4.69E-05	0.161	2.90E-04
H003367	2010	13	3.42E-04	0.150	2.27E-03
H003704	2010	14	1.27E-02	0.390	3.25E-02
R610785	2010	15	3.44E-04	0.020	1.76E-02
R610826	2010	16	3.57E-07	0.015	2.31E-05
R610853	2010	17	4.24E-06	0.061	6.91E-05
S001721	2010	18	4.70E-06	0.419	1.12E-05
H003443	2011	1	2.50E-02	0.411	6.08E-02
S002129	2011	2	4.57E-06	0.631	7.24E-06
H002592	2011	3	4.85E-06	0.023	2.09E-04
H003337	2011	4	4.32E-06	0.149	2.89E-05
S001105	2011	5	4.43E-06	0.526	8.43E-06
H003343	2011	6	4.72E-06	0.183	2.58E-05
H003371	2011	7	7.05E-03	0.221	3.19E-02
H003526	2011	8	1.88E-04	0.182	1.03E-03
H003565	2011	9	3.67E-04	0.217	1.69E-03

3013 Container	FY	DE	H₂ observed (mole)	H₂ calculated using pressure equation (mole)	H₂ Fraction
R611131	2011	10	5.15E-07	0.029	1.75E-05
H003625	2011	11	2.26E-04	0.193	1.17E-03
L000178	2011	12	5.13E-06	0.009	5.68E-04
H003328	2011	13	1.25E-01	0.711	1.76E-01
H001209	2012	1	2.09E-04	0.122	1.72E-03
H002574	2012	2	4.84E-07	0.149	3.25E-06
H001513	2012	3	4.87E-05	0.141	3.45E-04
H003390	2012	4	8.34E-05	0.198	4.22E-04
L000075	2012	5	5.24E-06	0.001	3.89E-03
H004012	2012	6	3.88E-05	0.200	1.94E-04
H004048	2012	7	1.36E-03	0.137	9.90E-03
R610960	2012	8	5.00E-03	0.061	8.19E-02
S002250	2012	10	4.86E-06	0.058	8.39E-05
H001236	2013	1	4.23E-04	0.164	2.58E-03
R610996	2014	1	3.50E-05	0.022	1.58E-03
H003064	2014	2	3.68E-02	0.414	8.87E-02
H003307	2014	3	4.51E-05	0.214	2.11E-04
H003052	2014	4	3.30E-03	0.270	1.22E-02
H003898	2014	5	7.65E-03	0.219	3.49E-02
S002277	2014	6	4.62E-06	0.593	7.80E-06
S002116	2014	7	4.89E-07	0.354	1.38E-06
H004219	2014	8	3.96E-04	0.183	2.16E-03
H002636	2014	9	4.96E-06	0.014	3.63E-04
R610156	2015	1	5.86E-07	0.219	2.67E-06
S002162	2015	2	4.64E-06	0.254	1.82E-05
H001979	2015	3	4.90E-06	0.128	3.83E-05
H001181	2015	4	6.43E-05	0.165	3.90E-04
H003181	2015	5	5.79E-03	0.394	1.47E-02
H003258	2015	6	4.65E-05	0.203	2.30E-04
H003737	2015	7	3.85E-03	0.331	1.16E-02
H003896	2015	8	4.06E-02	0.399	1.02E-01
H004302	2015	9	1.07E-02	0.259	4.12E-02
H001191	2016	1	3.47E-03	0.169	2.06E-02
H002556	2016	2	8.76E-05	0.210	4.18E-04
H004173	2016	3	2.91E-04	0.234	1.24E-03
H004247	2016	4	4.77E-06	0.180	2.65E-05
H003775	2016	5	4.33E-05	0.204	2.12E-04

3013 Container	FY	DE	H₂ observed (mole)	H₂ calculated using pressure equation (mole)	H₂ Fraction
H004024	2016	6	4.51E-05	0.256	1.76E-04
H001304	2017	1	1.68E-03	0.153	1.09E-02
H002575	2017	2	1.27E-02	0.409	3.10E-02
H003352	2017	3	8.89E-04	0.393	2.26E-03
H003695	2017	4	3.54E-02	0.368	9.64E-02
H002508	2017	5	1.87E-02	0.330	5.65E-02
R600793	2017	6	4.88E-06	0.803	6.08E-06

Appendix 7. Estimating bounds for the H₂ fraction in the 3013 container population from the DE observations.

Two statistical approaches were considered for determining upper bounds for the H₂ fraction – (1) nonparametric tolerance limits and (2) a distributional approach using the beta distribution. Both statistical approaches require that the sample data are representative of the entire population. These DE data are almost entirely from a random sample from the Pressure and Corrosion Bin container population. For this population, the random sampling provides confidence of the representativeness of the data. However, some of the containers were selected based on expert judgment, including HHMC. Thus, the sample could have some bias towards higher moisture content. These statistical approaches were used along with expert judgment to develop an expression for the maximum H₂ fraction for use in the 3013 pressure equation.

Nonparametric tolerance limits to assess bounds. The approach used to determine the nonparametric tolerance limits is described in Young and Mathew.¹⁹ A one-sided tolerance limit is a value such that there is a specified level of confidence (e.g., 90%, 95%, 99%, ...) that a percentage of the population (e.g., 95%, 97.5%, 99%, ...) is less than that value. To determine nonparametric tolerance limits requires very large sample sizes. To determine an upper 95%/97.5% tolerance limit requires 118 items using method described in Young and Mathew. There are 113 DE containers, however, one of these, the HHMC, is not compliant, therefore there are 112 compliant DE containers. Although not quite meeting the sample size requirement, extrapolation is reasonable in this case.¹⁹ The 95%/97.5% nonparametric estimate for the upper tolerance limit is 0.102. The 99%/97.5% estimate is 0.11. However, the sample size required for this estimate is 182, so this estimate could be underestimating the actual value. The software used in this analysis is the *tolerance* package in R.²⁰

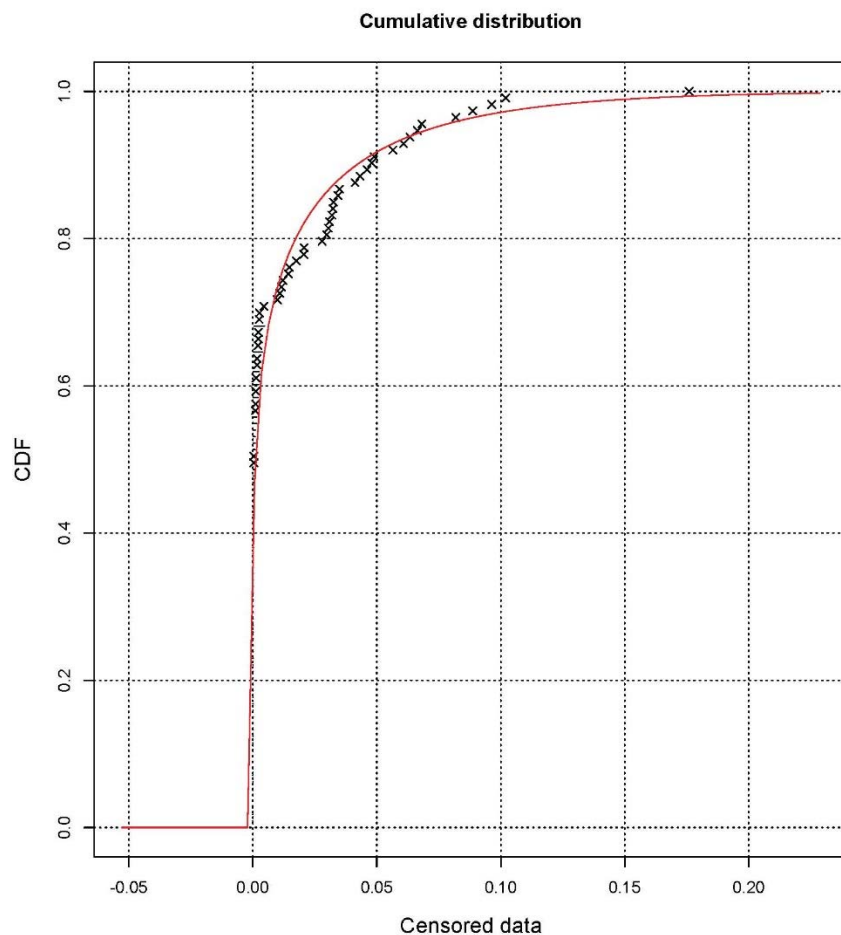
Distribution-based upper quantiles to assess bounds. Since the sample size is not adequate for determining all nonparametric tolerance limits of interest, a distribution-based approach is also considered. Fitting a probability distribution to the data allows estimating upper quantiles of the distribution and uncertainties associated with those estimates. An upper *k*th% quantile is a value such that *k*% of the population is less than that value.

Of the 114 H₂ fraction values (113 compliant container values), there are 6 non-detects, 43 trace values and 18 values reported as less than 0.1%. This is a substantial portion of the data and needs to be included in the analysis. The non-detect data are in the interval 0 to 0.001%, the trace data are in the interval 0.001% to 0.01% and the less than 0.1% are in the interval 0.01% to 0.1%. This kind of interval-censored data is well-known in the statistical literature.²¹ The approach used to fit the data is based upon maximum likelihood as described in Klein and Moeschberger.^{21a} The software used in this analysis is the *fitdistrplus* package in R.^{20b, 22}

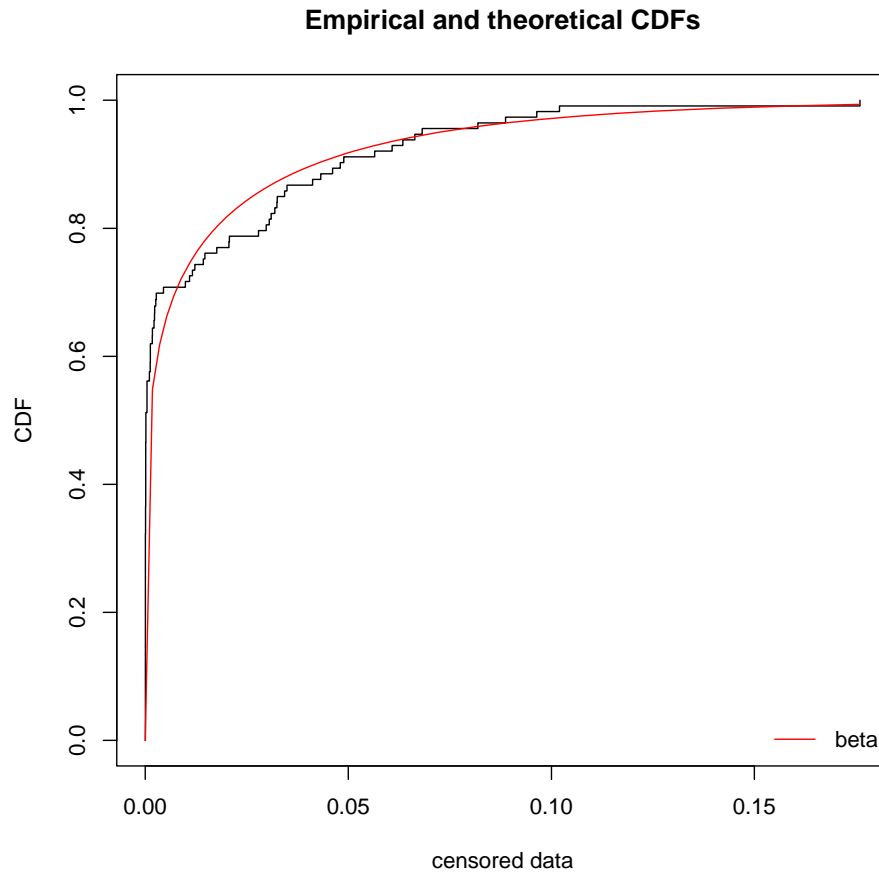
When choosing distributions for fitting data, one would like to have a physical justification for the choice. Although there are a number of mechanisms that result in the H₂ fraction behavior, the mechanisms are not well enough developed to be quantitative at this time. The H₂ fraction values are in the interval [0, 1] and it is reasonable to pick a distribution that is limited to this interval. The beta distribution is a two-parameter family of continuous probability distributions defined on the interval [0, 1] and has been applied to model random variables in a wide variety

of disciplines. The beta distribution is used in this analysis to fit both the total H₂ fraction data (114 containers) and the compliant H₂ fraction data (113 containers). Other distributions were considered (Gamma and Weibull), but neither of these provided a better fit when comparing cumulative probability distributions and other measures of goodness of fit (e.g. Akaike information criterion (AIC) and Bayesian information criterion (BIC)). In addition, they are not restricted to the [0,1] interval.

Appendix 7 Figure 1 shows the fit for all of the data, the small horizontal lines are the interval observations and the X's are the non-interval observations. The red curve is the beta cumulative distribution fitted to these data. As can be seen in the figure, the beta distribution provides a reasonable fit to the data. Appendix 7 Figure 2, compares the beta fit to the empirical distribution.¹⁹ This comparison indicates that the beta fit is good or perhaps even conservative in the upper tail (values greater than 0.08), which is the region of interest for this analysis.

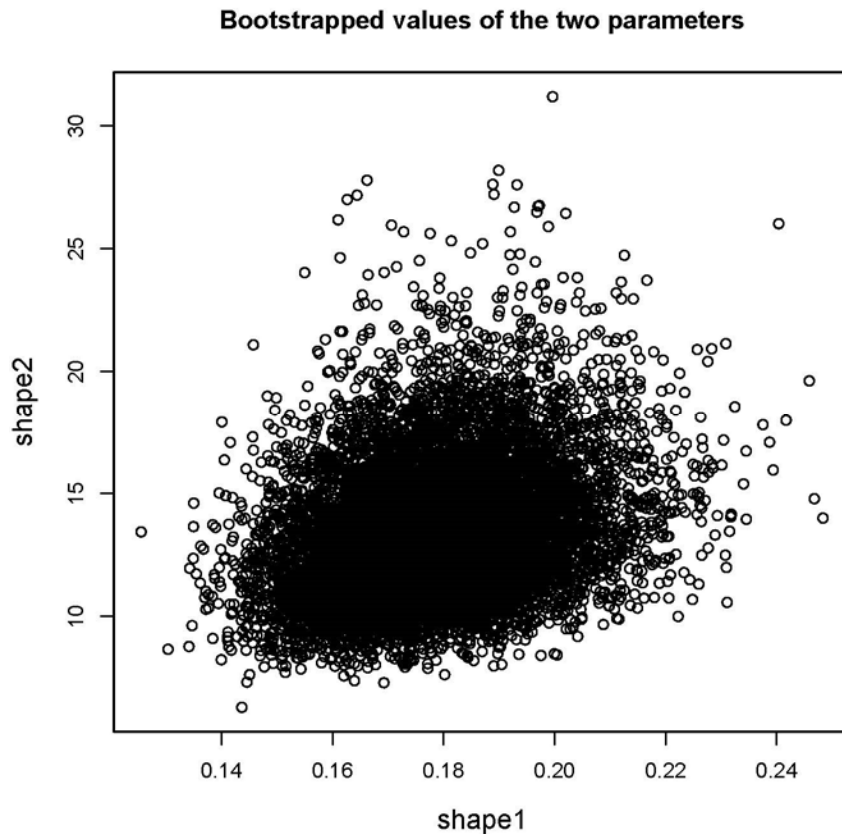


Appendix 7 Figure 1. The H₂ fraction data for 114 observations. The small horizontal lines are the interval observations and the X's are the non-interval observations. The red curve is the beta cumulative distribution fitted to these data.



Appendix 7 Figure 2. Comparison of the beta-fit cumulative distribution based on all of the H₂ Fraction data (red) to the empirical cumulative distribution (black).

To evaluate sampling uncertainties, the bootstrap method is used.²³ This is a simple, but powerful Monte Carlo method for determining confidence intervals and prediction uncertainties. The method is based on resampling with replacement from the original data, generating a large number of fits. Appendix 7 Figure 3 shows the two beta parameters (shape 1 and shape 2) for 10,000 resamples.



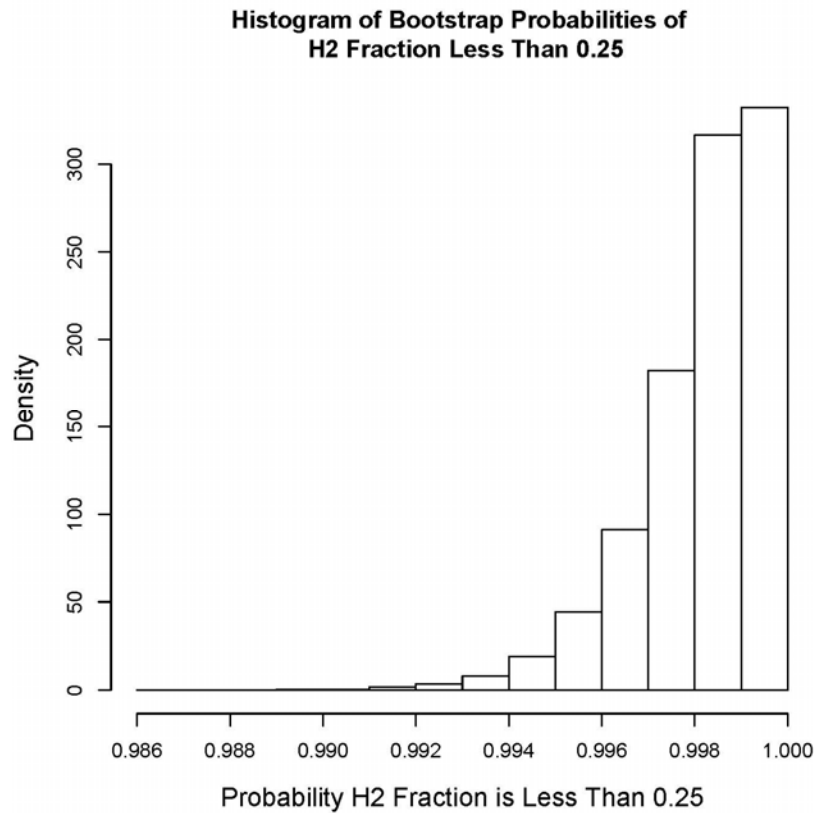
Appendix 7 Figure 3. Shows the shape parameters for 10,000 resampling fits to the original data set.

Appendix 7 Table 1 shows the 95%, 99% and 99.9% quantiles and the 95% upper confidence limit based on the bootstrap fits. For example, the value (quantile) such that 99% of the population is less than or equal to this value is 0.15 and the upper 95% confidence bound on this quantile is 0.20. When estimating upper quantiles the issue of model misspecification is particularly important. However, Modarres et al. recommend (based on simulation studies) that "when the size of the sample is large we should use the data to select a model, and then estimate the quantiles based on the selected model." In their paper large is a sample size of 100 or greater.²⁴

Appendix 7 Table 1. Quantiles and upper 95% confidence limits for 95%, 99%, 99.6% and 99.9% probabilities.

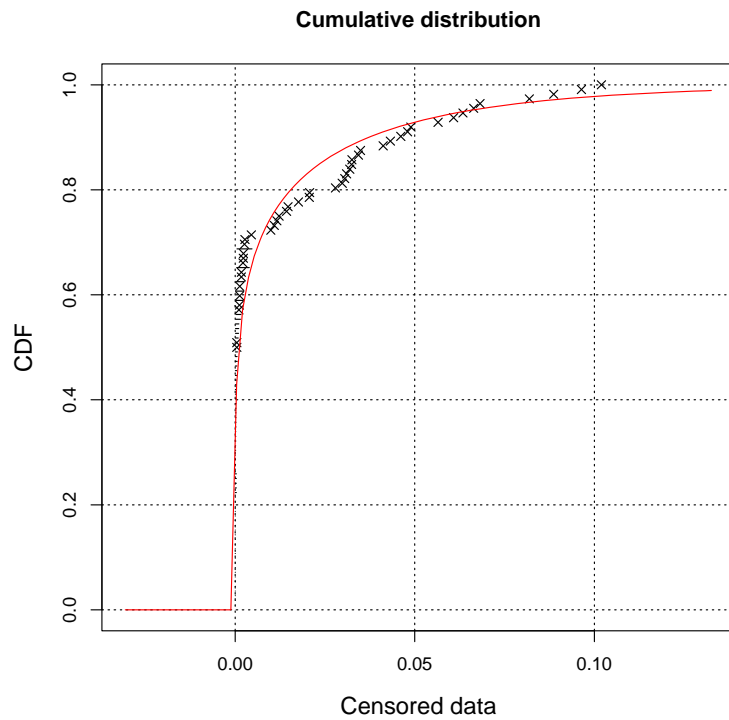
Probability	Quantile	95% Upper Confidence Limit
95%	0.07	0.095
99%	0.15	0.20
99.6%	0.20	0.26
99.9%	0.27	0.34

To explore the question of what is the probability and associated uncertainties that a future observation will be less than 0.25, these probabilities are determined for each of the 10000 beta fits. Appendix 7 Figure 4 shows a density histogram of those values. Denoting the probability that a future observation will be less than 0.25 as $P_{0.25}$, one can say that there is a 99.9% probability that $P_{0.25}$ is greater than or equal to 99.1%, at least 99.1% of the population is less than 0.25. These results combined with shelf life studies and expert judgment result in choosing a bound of 0.25 for the entire population (the endpoint of the green dot-dashed line in Figure 5).

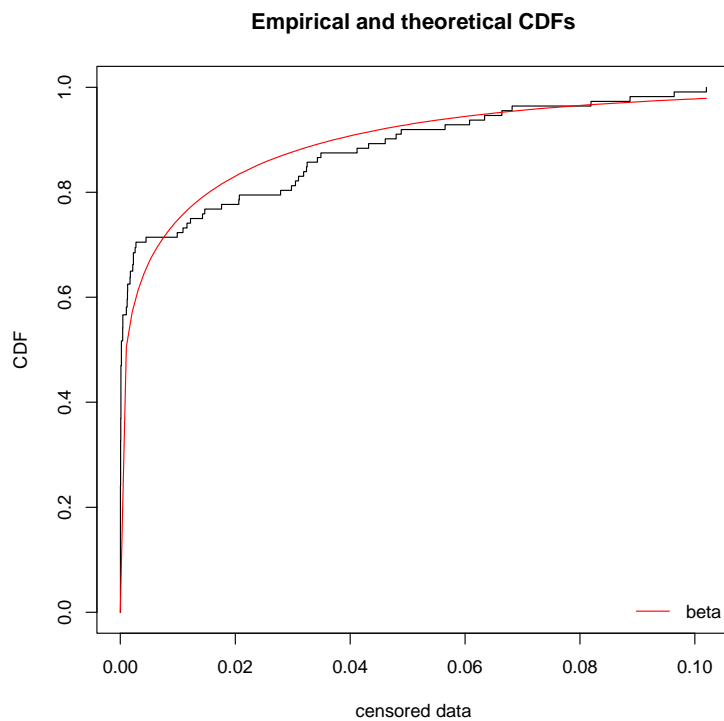


Appendix 7 Figure 4. Histogram of bootstrap probabilities of H₂ fraction less than 0.25.

The compliant population consists of all DEs except the HHMC. A beta distribution fit to the compliant population provides a reasonable fit to these data, Appendix 7 Figure 5. The results are given in Appendix 7 Table 2. It also appears to be slightly conservative for values greater than 0.08 based on the comparison of the beta-fit cumulative distribution to the empirical distribution shown in Appendix 7 Figure 6.



Appendix 7 Figure 5. The H₂ fraction data for 113 compliant observations. The small horizontal lines are the interval observations and the X's are the non-interval observations. The red curve is the beta cumulative distribution fitted to these data.



Appendix 7 Figure 6. Comparison of the beta-fit cumulative distribution based on the compliant H₂ Fraction data (red) to the empirical cumulative distribution (black).

The 97.5% quantile with 99% uncertainty is 0.13 (Appendix 7 Table 2), which is considerably larger than the nonparametric estimate of 0.102. This increases confidence that the bounding estimates based on the beta-distribution are conservative. These results combined with shelf-life studies and expert judgment result in choosing a bound of 0.13 (99% upper uncertainty limit for the 97.5% quantile **estimate**) for the H₂ Fraction for containers with moisture less than or equal to 0.31 wt% - the blue dashed line in Figure 5).

Appendix 7 Table 2. The results of a beta distribution fit of the H₂ fractions of the entire compliant population (HHMC excluded).

Quantile Percent	95% Uncertainty Limit for Quantile Estimate	99% Upper Uncertainty Limit for Quantile Estimate
97.5	0.12	0.13
99	0.17	0.19

References

1. Department of Energy, Stabilization, Packaging, and Storage of Plutonium-Bearing Materials. U.S. Department of Energy: Washington, D.C. 20585, 2012.
2. Farr, J. D.; Schulze, R. K.; Neu, M. P., Surface chemistry of Pu oxides. *Journal of Nuclear Materials* **2004**, 328, 124-136.
3. (a) Foy, B. R.; Joyce, S. A. *Gas-phase Radiolysis in Plutonium Dioxide Powder*; LA-UR-08-2627; Los Alamos National Laboratory: 2008; (b) Haschke, J. M.; Allen, T. H.; Morales, L. A., Reactions of plutonium dioxide with water and hydrogen–oxygen mixtures: Mechanisms for corrosion of uranium and plutonium. *Journal of Alloys and Compounds* **2001**, 314, 78-91.
4. Rios, D.; Gaunt, A. J.; Narlesky, J. E.; Berg, J. M.; Veirs, D. K.; Worl, L. A. *Capture, Identification and Quantification of the Elusive Chlorine-Containing Gases Emitted from Hydrated PuO₂/Salt Mixtures*; LA-UR-17-27871; Los Alamos National Laboratory: Los Alamos, NM, 2017.
5. Veirs, D. K.; Berg, J. M.; Worl, L. A.; Narlesky, J. E.; Dunn, K. A.; Louthan, J., M.R., Evidence of Corrosive Gas Formed by Radiolysis of Chloride Salts in Plutonium-bearing Materials. *Journal of Nuclear Material Management* **2010**, 38 (3), 25 - 31.
6. Veirs, D. K.; Berg, J. M. *Maximum hydrogen pressure predicted in MIS salt-bearing materials*; LA-UR-08-06289; Los Alamos National Laboratory: Los Alamos, NM, 2008.
7. (a) James, M. R.; Joyce, S. A.; Berg, J. M.; Veirs, D. K. *PuO₂-alpha modeling progress*; LA-UR-20422; Los Alamos National Laboratory: Los Alamos, NM, 2016; (b) Veirs, D. K. *Gas Generation and Radiation Chemistry*; LA-UR-16-21467; Los Alamos National Laboratory: Los Alamos, NM, 2016.
8. Berg, J. M.; Carillo, A.; Martinez, M. A.; Veirs, D. K.; Worl, L. A.; Harradine, D. M.; McInroy, R. E. *Predicting the future gas composition of MIS material 011589A*; LA-UR-07-7870; Los Alamos National Laboratory: Los Alamos, NM 87545, 2007.
9. Assistant Manager Nuclear Material Stabilization *Integrated Surveillance and Monitoring Program for Materials Packaged to Meet DOE-STD-3013*; AMNMS-15-0014; Department of Energy Savannah River: Aiken, SC, 2015.
10. Rawls, G. B. *KIS Can Puncture Device Design Modification and Performance Testing*; WSRC-TR-2007-00078; Westinghouse Savannah River Company: Aiken, SC, 2007.
11. Arnold, M. J. *FY08 3013 Surveillance Processing Plan for K-Area*; G-ESR-K-00014, Rev. 1; Savannah River Site: Aiken, SC, 2007.
12. (a) Hardy, B. J. *Version 2.0 of the 3013 Gas Evaluation Software Tool (GEST)*; WSRC-TR-2007-00218; Westinghouse Savannah River Aiken, SC, 2007; (b) Laurinat, J. E. *Version 2.3 of the 3013 Gas Evaluation Software Tool (GEST)*; G-TRT-A-00005, Rev. 3; Savannah River Site: Aiken, SC, 2014.
13. Arnold, M. J. *3013 Gas Composition Software Evaluation*; Y-ESR-K-00003, Rev. 0; Savannah River Site: Aiken, SC, 2008.
14. Laurinat, J. E. *Software Quality Assurance Plan for Version 2.3 of the 3013 Gas Evaluation Software Tool (GEST)*; G-SQP-A-00016 Rev. 3; Savannah River National Laboratory: Aiken, SC, 2014.
15. Duffey, J. M. *A Comparison of Gas Composition and Pressure of Inner and Outer 3013 DE Containers*; SRNL-STI-2014-00439; Savannah River National Laboratory: Aiken, SC, 2014.

16. Berg, J. M.; Crowder, M. L.; Almond, P. M., Relationship Between Reported, As-Packaged Moisture, and Moisture Measurements Made During Surveillance of 3013 Containers. *Journal of Nuclear Material Management* **2010**, 37 (3), 101-107.
17. Mulford, R. N. *Helium Transport in PuO₂ as a Function of Temperature and Heating Rate*; LA-UR-14-25325; Los Alamos National Laboratory: Los Alamos, NM, 2014.
18. Sims, H. E.; Webb, K. J.; Brown, J.; Morris, D. E.; Taylor, R. J., Hydrogen yields from water on the surface of plutonium dioxide. *Journal of Nuclear Materials* **2013**, 437, 359-364.
19. Young, D. S.; Mathew, T., Improved nonparametric tolerance intervals based on interpolated and extrapolated order statistics. *Journal of Nonparametric Statistics* **2014**, 26 (3), 415-432.
20. (a) Young, D. S., Tolerance: An R Package for Estimating Tolerance Intervals. *Journal of Statistical Software* **2010**, 36 (5), 1-39; (b) R Core Team R: A language and environment for statistical computing. <http://www.R-project.org/>.
21. (a) Klein, J. P.; Moeschberger, M. L., *Survival Analysis: Techniques for Censored and Truncated Data*. 2nd ed.; Springer-Verlag: 2003; (b) Helsel, D. R., *Nondetects and Data Analysis: Statistics for Censored Environmental Data*. John Wiley & Sons: 2005.
22. Delignette-Muller; Laure, M.; Dugang, C., fitdistrplus: An R Package for Fitting Distributions. *Journal of Statistical Software* **2015**, 64 (4), 1-34.
23. Efron, B.; Tibshirani, R. J., *An Introduction to the Bootstrap*. 1st Edition ed.; Chapman & Hall: 1994.
24. Modarres, R.; Nayak, T. K.; Gastwirth, J. L., Estimation of upper quantiles under model and parameter uncertainty. *Computational Statistics & Data Analysis* **2002**, 39 (4), 529-554.